

An Intrinsic Topological Invariant in Strongly Interacting Quantum Systems

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It is still an outstanding challenge to characterize and understand the topological features of strongly correlated states such as bound-states in interacting quantum systems. Here, from the cotranslational symmetry in an interacting multi-particle quantum system, we develop a general method to construct an intrinsic Chern invariant for identifying strongly correlated topological states. As an example, we study the topological magnons in a strongly interacting two-dimensional spinor Hofstadter model, which can be realized by the currently experimental techniques [Phys. Rev. Lett. **111**, 185301 (2013); Phys. Rev. Lett. **111**, 185302 (2013)]. Through calculating the two-magnon excitation spectrum and the intrinsic Chern number, we explore the emergence of topological edge bound-states and give their topological phase diagram. We also analytically derive an effective single-particle Hofstadter superlattice model for understanding the topological bound-states. Our results not only provide a new approach to defining topological invariants, but also give deep insights into the characterization and understanding of strongly correlated topological states.

Topological invariants, which describe the invariant property of a topological space under homeomorphisms, are of great importance in characterizing topological matters and topological phase transitions. Weakly interacting topological states, whose universal properties do not depend on inter-particle interactions, are well-understood due to the well-developed tools for treating weakly interacting systems [1–4]. However, strongly interacting topological states, whose universal properties are determined by inter-particle interactions, pose much greater challenges to both theory [5] and experiment [6]. The characterization of strongly interacting topological states is quite different from that of the weakly interacting counterparts [7, 8]. Due to the existence of strong correlations among particles, it is hard to define a topological invariant and to clarify the interplay between topological features and inter-particle interactions.

Ultracold atoms in optical lattices offer a well-controlled experimental platform to explore topological matters in a clean environment [9]. Recently, the Hofstadter-Harper model has been experimentally realized by using laser-assisted tunneling of ultracold atoms in a tilted optical potential [10, 11]. As the atom-atom interaction can be tuned by Feshbach resonances, such an atomic Hofstadter-Harper system not only opens a way to explore topological states of noninteracting atoms, but also provides new opportunity to study strongly correlated topological states.

Beyond single-particle topological states [12–18], it is of great challenge to clarify whether correlated topological states may emerge. One outstanding challenge is the absence of a well-defined topological invariant for an interacting quantum system (IQS). In this Letter, we find that this problem can be solved when the system has *cotranslational symmetry*: the invariance under collective translation. We demonstrate that the cotranslational

symmetry naturally allows us to formulate a topological invariant, which can be used to characterize the topological features of correlated multi-particle states such as bound-states (BS's). In comparison with other topological invariants, our topological invariant is intrinsic and straightforward. A well-known generalization of the Thouless-Kohmoto-Nightingale-den Nijs (TKNN) invariant [19] from noninteracting to interacting systems is by introducing the twisted boundary condition (BC) [20], which requires to calculate all many-body ground-states for a continuous 2π -period of the twist angle. Another topological invariant for IQS's is given in terms of the Green's function, which requires to calculate the Green's function at all frequencies [21] or zero frequency [22]. Differently, our topological invariant is directly defined by using the center-of-mass (c.o.m) quasi-momentum associated with the cotranslational symmetry. We believe that our definition opens a new route to the characterization of strongly correlated topological states.

Topological invariant associated with cotranslational symmetry. – To illustrate our idea, we consider a generally two-dimensional (2D) quantum system with N interacting particles. The Hamiltonian reads as,

$$H = \sum_{j=1}^N H_j + \sum_{j=1}^{N-1} \sum_{j'=j+1}^N V(|\mathbf{r}_j - \mathbf{r}_{j'}|). \quad (1)$$

Here, $\mathbf{r}_j = (x_j, y_j)$ is the position of the j -th particle, the single-particle Hamiltonian H_j is of translational symmetry with respect to the period $\mathbf{a} = (a_x, a_y)$, and the interaction $V(|\mathbf{r}_j - \mathbf{r}_{j'}|)$ only depends on the inter-particle distance. Typical examples are quantum lattice models such as Hubbard lattices and quantum spin lattices. Although we concentrate on quantum lattice models, our idea can be extended to continuous models.

Given an N -particle wave-function $\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$, the single-particle translation operator for the j -th par-

ticle, $T^{(j)}$, is defined as $T^{(j)}\psi(\mathbf{r}_1, \dots, \mathbf{r}_j, \dots, \mathbf{r}_N) = \psi(\mathbf{r}_1, \dots, \mathbf{r}_j + \mathbf{a}, \dots, \mathbf{r}_N)$ with $j \in \{1, 2, \dots, N\}$. For a noninteracting system, because of the translational symmetry of each single-particle Hamiltonian H_j , $T^{(j)}$ commutes with the whole Hamiltonian and the many-body eigenstate has a tensor product structure of N single-particle Bloch states. Therefore the independent Bloch momenta of the N particles form a set of good quantum numbers for the noninteracting Hamiltonian. However, the interaction will break the single-particle translational symmetry and make the N independent Bloch momenta no longer good quantum numbers.

We now define the cotranslation operator, $T_a(\tau)$, as $T_a(\tau)\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = \psi(\mathbf{r}_1 + \tau\mathbf{a}, \mathbf{r}_2 + \tau\mathbf{a}, \dots, \mathbf{r}_N + \tau\mathbf{a})$ with τ an arbitrary integer. Actually, $T_a(\tau)$ is a combination of all single-particle translation operators, $T_a(\tau) = [T^{(1)}T^{(2)}\dots T^{(N)}]^\tau$, and thus it commutes with each H_j . Since $T_a(\tau)T_a(\tau') = T_a(\tau')T_a(\tau) = T_a(\tau + \tau')$ and $[T_a(\tau)]^{-1} = T_a(-\tau)$, the set $\{T_a(\tau), \tau \in \mathbb{Z}\}$ forms an Abelian group (where \mathbb{Z} is the set of all integers). We call this group as the *cotranslation group*. As all cotranslation operators commute with the interaction term, the whole Hamiltonian is invariant under the cotranslation transform, i.e. $[T_a(\tau)]^{-1}HT_a(\tau) = H$, which represents the *cotranslational symmetry*.

Under the cotranslational symmetry, the Hamiltonian H and $T_a(\tau)$ share a set of common eigenstates. The common eigenstates obey

$$T_a(\tau)\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = c_a(\tau)\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N), \quad (2)$$

with $c_a(\tau)$ being an eigenvalue of $T_a(\tau)$. It is easy to find $c_a(\tau)c_a(\tau') = c_a(\tau + \tau')$ and $[c_a(\tau)]^{-1} = c_a(-\tau)$. Thus the eigenvalues could be chosen as the exponential form $c_a(\tau) = e^{i\mathbf{k} \cdot \tau\mathbf{a}}$ with the vector $\mathbf{k} = (k_x, k_y)$ [23], which is a pair of good quantum numbers. Thus we have,

$$\psi(\mathbf{r}_1 + \tau\mathbf{a}, \dots, \mathbf{r}_N + \tau\mathbf{a}) = e^{i\mathbf{k} \cdot \tau\mathbf{a}}\psi(\mathbf{r}_1, \dots, \mathbf{r}_N), \quad (3)$$

which resembles the Bloch theorem for single-particle systems with translational symmetry. Therefore, the vector \mathbf{k} acts as the corresponding c.o.m quasi-momentum. Similar to the Bloch functions for single-particle systems with translational symmetry, one can define $\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = e^{i\mathbf{k} \cdot \frac{1}{N}(\mathbf{r}_1 + \mathbf{r}_2 + \dots + \mathbf{r}_N)}\phi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$ and then obtain $\phi(\mathbf{r}_1 + \tau\mathbf{a}, \mathbf{r}_2 + \tau\mathbf{a}, \dots, \mathbf{r}_N + \tau\mathbf{a}) = \phi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$ from Eq. (3). We thus identify these eigenstates $\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$ as many-body Bloch states for IQS's with cotranslational symmetry.

By exploiting the cotranslational symmetry and the many-body Bloch states, we define a topological invariant (the first Chern number). It is an integral of the Berry curvature $\mathcal{F}_n(k_x, k_y)$ over the first Brillouin zone (BZ),

$$C_n = \frac{1}{2\pi} \iint_{\text{BZ}} d^2\mathbf{k} \mathcal{F}_n(k_x, k_y), \quad (4)$$

where, $\mathcal{F}_n(k_x, k_y) = \text{Im} \left(\left\langle \frac{\partial \phi_n}{\partial k_x} \left| \frac{\partial \phi_n}{\partial k_y} \right\rangle - \left\langle \frac{\partial \phi_n}{\partial k_y} \left| \frac{\partial \phi_n}{\partial k_x} \right\rangle \right) \right)$ is determined by the Bloch state $|\phi_n\rangle = |\phi_n(k_x, k_y)\rangle$, $k_x \in$

$(-\pi/a_x, \pi/a_x]$, $k_y \in (-\pi/a_y, \pi/a_y]$, and n is the band index. In fact, the above Chern number is a TKNN-type topological invariant. We should remark that our topological invariant is always well-defined for the band which is well-separated from other bands, that is, it is protected by the corresponding energy gaps.

Strongly interacting spinor Hofstadter model. – We consider an ensemble of two-component Bose atoms in a 2D optical lattice subjected to artificial magnetic fields. The system obeys an interacting spinor Hofstadter model (see Supplementary Material [24]),

$$\begin{aligned} \hat{H}_B = & - \sum_{l,m,\sigma} [K e^{i\alpha_\sigma m\Phi} \hat{a}_{l+1,m,\sigma}^\dagger \hat{a}_{l,m,\sigma} + \text{h.c.}] \\ & - \sum_{l,m,\sigma} [J \hat{a}_{l,m+1,\sigma}^\dagger \hat{a}_{l,m,\sigma} + \text{h.c.}] \\ & + \sum_{l,m,\sigma_1,\sigma_2} \frac{1}{2} U_{\sigma_1\sigma_2} \hat{n}_{l,m,\sigma_1} (\hat{n}_{l,m,\sigma_2} - \delta_{\sigma_2}^{\sigma_1}), \end{aligned} \quad (5)$$

with the lattice index $\{l, m\}$, the component index $\sigma \in \{\uparrow, \downarrow\}$, the creation (annihilation) operators $\hat{a}_{l,m,\sigma}^\dagger$ ($\hat{a}_{l,m,\sigma}$), the number operator $\hat{n}_{l,m,\sigma} = \hat{a}_{l,m,\sigma}^\dagger \hat{a}_{l,m,\sigma}$, and the Kronecker delta $\delta_{\sigma_2}^{\sigma_1}$. Here, $U_{\sigma_1\sigma_2}$ is the on-site interaction, and K and J are respectively the hopping strengths along x and y directions. The hopping along x -direction involves an additionally spin-spatial-dependent phase $\phi_{\sigma,m} = \alpha_\sigma m\Phi$ with $\alpha_\uparrow = 1$ and $\alpha_\downarrow = -1$.

In the strong interaction regime with unit filling, by using the second-order perturbation theory [25], one can map the model (5) onto a 2D Heisenberg spin model, $\hat{H}_H = -J_x \sum_{l,m} [(e^{i2m\Phi} \hat{S}_{l+1,m}^+ \hat{S}_{l,m}^- + \lambda \hat{S}_{l,m+1}^+ \hat{S}_{l,m}^-) + \text{h.c.}] - V_x \sum_{l,m} [\hat{S}_{l,m}^z \hat{S}_{l+1,m}^z + \lambda \hat{S}_{l,m}^z \hat{S}_{l,m+1}^z]$ (see Supplementary Material [24]). Here, $\hat{S}_{l,m}^+ = \hat{a}_{l,m,\uparrow}^\dagger \hat{a}_{l,m,\downarrow}$, $\hat{S}_{l,m}^- = \hat{a}_{l,m,\downarrow}^\dagger \hat{a}_{l,m,\uparrow}$, and $\hat{S}_{l,m}^z = \frac{1}{2}(\hat{n}_{l,m,\uparrow} - \hat{n}_{l,m,\downarrow})$. The parameters are given as $J_x = 2K^2/U_{\uparrow\downarrow}$, $J_y = \lambda J_x$, $V_x = 4K^2(1/U_{\uparrow\uparrow} + 1/U_{\downarrow\downarrow} - 1/U_{\uparrow\downarrow})$, $V_y = \lambda V_x$, and $\lambda = V_y/V_x = J^2/K^2$. Different from the system without gauge fields, our \hat{H}_H includes a spatially varying phase $2m\Phi$ along x -direction. According to the Matsubara-Matsuda mapping [26], the model \hat{H}_H is equivalent to a hard-core Bose-Hubbard model. By introducing $|\downarrow\rangle \leftrightarrow |0\rangle$, $|\uparrow\rangle \leftrightarrow |1\rangle$, $\hat{S}_{l,m}^+ \leftrightarrow \hat{b}_{l,m}^\dagger$, $\hat{S}_{l,m}^- \leftrightarrow \hat{b}_{l,m}$, and $\hat{S}_{l,m}^z \leftrightarrow \hat{b}_{l,m}^\dagger \hat{b}_{l,m} - \frac{1}{2}$, we have,

$$\begin{aligned} \hat{H} = & -J_x \sum_{l,m} (e^{i2\pi\beta m} \hat{b}_{l+1,m}^\dagger \hat{b}_{l,m} + \lambda \hat{b}_{l,m+1}^\dagger \hat{b}_{l,m} + \text{h.c.}) \\ & - V_x \sum_{l,m} (\hat{n}_{l,m} \hat{n}_{l+1,m} + \lambda \hat{n}_{l,m} \hat{n}_{l,m+1}) \end{aligned} \quad (6)$$

with the hard-core bosonic creation (annihilation) operators $\hat{b}_{l,m}^\dagger$ ($\hat{b}_{l,m}$) and the number operator $\hat{n}_{l,m} = \hat{b}_{l,m}^\dagger \hat{b}_{l,m}$. Here, $\beta = \Phi/\pi$ and we remove a constant energy shift. Below, we concentrate on discussing the rational flux $\beta = p/q$ (where p and q are coprime integers).

Topological two-magnon excitations. – The two-particle Hilbert space can be spanned by the basis $|l_1, m_1; l_2, m_2\rangle = \hat{b}_{l_1, m_1}^\dagger \hat{b}_{l_2, m_2}^\dagger |\mathbf{0}\rangle$. An arbitrary two-particle state can be expressed as $|\Psi\rangle = \sum_{l_1, m_1; l_2, m_2} \psi_{l_1, m_1; l_2, m_2} |l_1, m_1; l_2, m_2\rangle$ with $\psi_{l_1, m_1; l_2, m_2} = \langle \mathbf{0} | \hat{b}_{l_2, m_2} \hat{b}_{l_1, m_1} | \Psi \rangle$. We consider a lattice of $L_x \times L_y$ sites and $L_y = qs$ with an odd integer s . Define the cotranslation operator, $T_{1,q} \psi_{l_1, m_1; l_2, m_2} = \psi_{l_1+1, m_1+q; l_2+1, m_2+q}$, the Hamiltonian (6) is invariant under $T_{1,q}$ (see Supplementary Material [24]). The two-particle Bloch states can be written as $\psi_{l_1, m_1; l_2, m_2} = e^{ik_x \frac{1}{2}(l_1+l_2) + ik_y \frac{1}{2}(m_1+m_2)} \varphi_{l_1, m_1; l_2, m_2}$ with $\varphi_{l_1+1, m_1+q; l_2+1, m_2+q} = \varphi_{l_1, m_1; l_2, m_2} = \varphi_{l_1, m_1+q; l_2, m_2+q}$. Here, $k_x = 2\pi\alpha_x/L_x$ with integer $\alpha_x \in [-\frac{L_x-1}{2}, \frac{L_x-1}{2}]$, $k_y = 2\pi\alpha_y/L_y$ with integer $\alpha_y \in [-\frac{s-1}{2}, \frac{s-1}{2}]$.

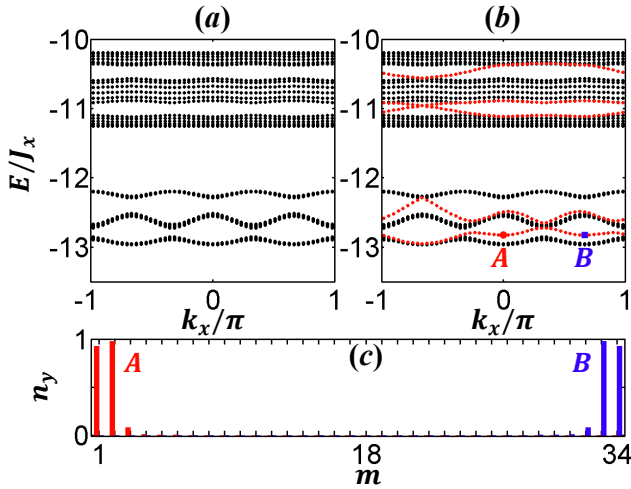


FIG. 1. (color online). Two-magnon bound-state spectra. Bound-state bands for (a) periodic BC along y -direction with $L_y = 33$ and (b) open BC along y -direction with $L_y = 34$. Both (a) and (b) choose the periodic BC along x -direction with $L_x = 51$. (c) The density distribution along y -direction for A (red point) with $k_x = 0$ and B (blue point) with $k_x = 2\pi/3$ in (b). The other parameters are chosen as $\beta = 1/3$, $V_x/J_x = 10$ and $\lambda = 1.2$.

We now discuss the energy spectrum. Under strong interactions, in addition to the continuum band, there appear BS bands. In Fig. 1(a), we show the BS spectrum under periodic BCs. It includes 6 subbands for $\beta = 1/3$ (in general, it includes $2q$ subbands for $\beta = p/q$). Based on our calculation, the Chern numbers for the 6 subbands are $(C_1, C_2, \dots, C_6) = (-1, 2, -1, -1, 2, -1)$. The Chern numbers indicate the bulk system has a non-trivial topology. According to the bulk-edge correspondence, topological edge states will appear in the system under open BC. So we calculate the spectrum under the open BC along y -direction. In Fig. 1(b), in addition to the extended BS's, topological edge BS's do appear. In Fig. 1(c), corresponding to the two points (A, B) in Fig. 1(b), we show their density distributions along y -

direction [$n_y(m) = \langle \hat{n}_y(m) \rangle = \langle \sum_l \hat{n}_{l,m} \rangle$]. The density distributions clearly show that these BS's do localize on the edges.

Topological phase transitions (TPTs). – The interaction ratio λ plays an important role in the BS spectrum. If $\lambda \gg 1$ (i.e. the interaction along y -direction dominates), the eigenstates of the three lowest subbands can be approximated by a superposition of $|l, m; l, m+1\rangle$, which are called y -type BS's. While the eigenstates of the three higher subbands can be approximated by a superposition of $|l, m; l+1, m\rangle$, which are called x -type BS's. If $\lambda \approx 1$ (i.e. $V_x \approx V_y$), the BS's are approximated by superpositions of $|l, m; l+1, m\rangle$ and $|l, m; l, m+1\rangle$. Otherwise, if $\lambda \ll 1$, the three lowest subbands correspond to x -type BS's while three higher subbands correspond to y -type BS's. By introducing $P_x = \sum_{l,m} |\psi_{l,m;l+1,m}|^2$ and $P_y = \sum_{l,m} |\psi_{l,m;l,m+1}|^2$, we have $P_x = 1$ ($P_y = 1$) for a perfect x -type (y -type) BS. For an arbitrary BS, we find that $P = P_x + P_y \simeq 1$ (see Supplementary Material [24]).

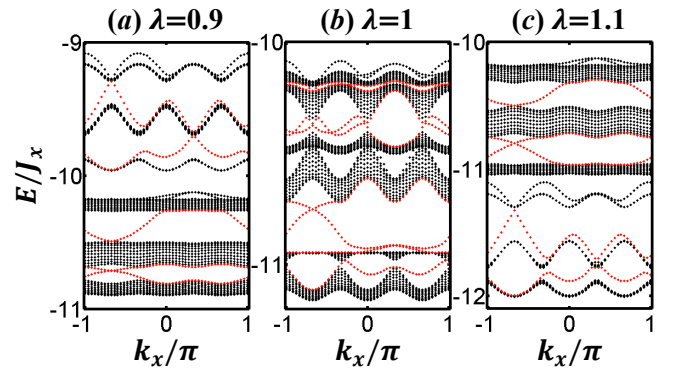


FIG. 2. Bound-state spectra with open BC along y direction and (a) $\lambda = 0.9$, (b) $\lambda = 1$ and (c) $\lambda = 1.1$. The other parameters are chosen as $\beta = 1/3$, $V_x/J_x = 10$, $L_x = 51$, and $L_y = 34$.

In Fig. 2, we show the BS spectra for different λ . Given the Riemann surface of Bloch states, the energy gaps represent the holes in the Riemann surface and the winding number of the edge states around these holes is another topological invariant [27, 28]. We find the absolute value of the winding number W_1 for the edge states in the first energy gap: $|W_1| = 1$ for $\lambda = 0.9$ and 1.1 [see Fig. 2(a,c)] and $|W_1| = 2$ for $\lambda = 1$ [see Fig. 2(b)]. This means that TPTs appear in the two regions: $0.9 < \lambda < 1$ and $1 < \lambda < 1.1$. Our calculations show that the Chern numbers for the lowest subband are $C_1 = (-1, 2, -1)$ for $\lambda = (0.9, 1, 1.1)$, which are consistent with the winding numbers for the corresponding edge states.

According to the topological band theory [1, 2], TPTs associate with gap closures. For a finite system, a gap closure corresponds to a gap minimum which approaches to zero when the system size increases. In Fig. 3, we show the topological phase diagram for the first BS

subband. Obviously, one can observe TPTs by varying λ and V_x/J_x , which are respectively determined by the hopping ratio J/K and the two interaction ratios ($U_{\uparrow\downarrow}/U_{\uparrow\uparrow}, U_{\uparrow\downarrow}/U_{\downarrow\downarrow}$) of the spinor Hofstadter model (5).

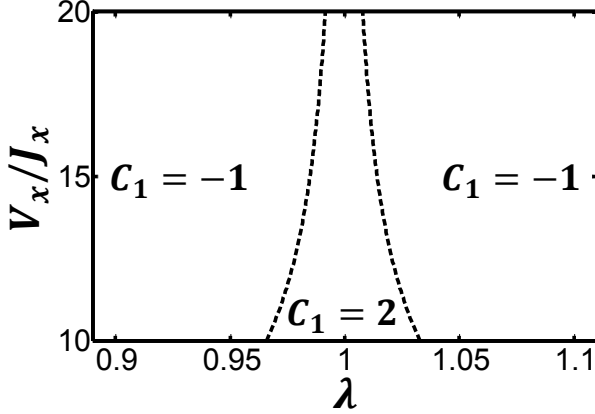


FIG. 3. Topological phase diagram for the lowest bound-state subband under periodic BCs with $\beta = 1/3$.

Effective single-particle Hofstadter superlattices. – Under strong interactions ($|J_x/V_x| \ll 1$), a BS can be regarded as a quasiparticle. By treating the hopping as a perturbation to the interaction and implementing the Schrieffer-Wolff transformation [29], the system obeys an effective single-particle model (see Supplementary Material [24]),

$$\begin{aligned} \hat{H}_{\text{eff}} &= \hat{H}_A + \hat{H}_B + \hat{H}_{AB}, \\ \hat{H}_A &= -J_{\text{eff}} \sum_{l,m} [(e^{i4\pi\beta m} \hat{A}_{l+1,m}^\dagger \hat{A}_{l,m} + \text{h.c.}) \\ &\quad + 2\lambda^2 (\hat{A}_{l,m+1}^\dagger \hat{A}_{l,m} + \text{h.c.}) + \epsilon_x \hat{A}_{l,m}^\dagger \hat{A}_{l,m}], \\ \hat{H}_B &= -J_{\text{eff}} \sum_{l,m} [\frac{2}{\lambda} (e^{i4\pi\beta m} e^{i2\pi\beta} \hat{B}_{l+1,m}^\dagger \hat{B}_{l,m} + \text{h.c.}) \\ &\quad + \lambda (\hat{B}_{l,m+1}^\dagger \hat{B}_{l,m} + \text{h.c.}) + \epsilon_y \hat{B}_{l,m}^\dagger \hat{B}_{l,m}], \\ \hat{H}_{AB} &= -J_{\text{eff}} J_{xy} \sum_{l,m} [e^{i2\pi\beta m} e^{i\pi\beta} (\hat{A}_{l,m}^\dagger \hat{B}_{l,m} + \hat{B}_{l+1,m}^\dagger \hat{A}_{l,m} \\ &\quad + \hat{A}_{l,m+1}^\dagger \hat{B}_{l,m} + \hat{B}_{l+1,m}^\dagger \hat{A}_{l,m+1}) + \text{h.c.}]. \end{aligned} \quad (7)$$

Here, $J_{\text{eff}} = J_x^2/V_x$, $J_{xy} = (\lambda + 1) \cos(\pi\beta)$, $\epsilon_x = V_x^2/J_x^2 + 2 + 4\lambda^2$, and $\epsilon_y = \lambda V_x^2/J_x^2 + 2\lambda + 4/\lambda$. The operators $\hat{A}_{l,m}^\dagger$ and $\hat{B}_{l,m}^\dagger$ create a particle in states $|l, m; l+1, m\rangle$ and $|l, m; l, m+1\rangle$, respectively. In Fig. 4(a), we show the lattice structure, in which the green and red circles respectively represent the sublattice-A and B. Actually, \hat{H}_A and \hat{H}_B are standard Hofstadter Hamiltonians, and \hat{H}_{AB} describes the coupling between the two sublattices.

Now we discuss the spectrum. Under the periodic BC along x -direction, through the Fourier transformation: $\hat{A}_{l,m}^\dagger = \frac{1}{\sqrt{L_x}} \sum_{k_x} e^{-ik_x l} \hat{A}_{k_x,m}^\dagger$ and $\hat{B}_{l,m}^\dagger = \frac{1}{\sqrt{L_x}} \sum_{k_x} e^{-ik_x l} \hat{B}_{k_x,m}^\dagger$, the system (7) becomes block di-

agonalized. The eigenstates $|\Psi(k_x)\rangle = [\psi_m^A(k_x) \hat{A}_{k_x,m}^\dagger + \psi_m^B(k_x) \hat{B}_{k_x,m}^\dagger] |\mathbf{0}\rangle$ obey the coupled Harper equations,

$$\begin{aligned} E' \psi_m &= - \begin{bmatrix} J^A & J_{m-1} e^{i\frac{k_x}{2}} \\ 0 & J^B \end{bmatrix} \psi_{m-1} - \begin{bmatrix} J^A & 0 \\ J_m e^{-i\frac{k_x}{2}} & J^B \end{bmatrix} \psi_{m+1} \\ &\quad - \begin{bmatrix} \epsilon_m^A & J_m e^{i\frac{k_x}{2}} \\ J_m e^{-i\frac{k_x}{2}} & \epsilon_m^B \end{bmatrix} \psi_m, \end{aligned} \quad (8)$$

with $E' = E/J_{\text{eff}}$, $\psi_m = [\psi_m^A, \psi_m^B]^T$, $J^A = 2\lambda^2$, $J^B = \lambda$, $\epsilon_m^A = \epsilon_x + 2 \cos(4\pi\beta m - k_x)$, $\epsilon_m^B = \epsilon_y + (4/\lambda) \cos(4\pi\beta m + 2\pi\beta - k_x)$, and $J_m = 2J_{xy} \cos(2\pi\beta m + \pi\beta - k_x/2)$. In Fig. 4 (b, c, d), we show the spectrum versus β . At $\lambda = 0.8$ and 1.2 , the butterfly-like spectrum includes two separated parts. When $\lambda \rightarrow 1$, the gap between the two parts gradually vanishes. Finally, at $\lambda = 1$, the two parts merge into one butterfly. Actually, such a spectrum deformation can be induced by tuning the hopping ratio J/K of the spinor Hofstadter model (5).

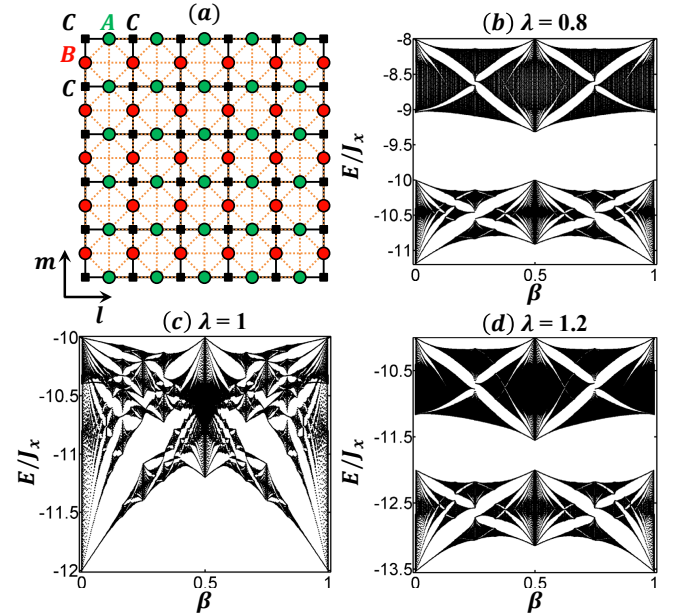


FIG. 4. (color online). Hofstadter superlattice and its butterfly-like spectra. (a) The sublattices A and B are respectively denoted by green and red circles. The original lattice C is represented by black squares. The sublattice site $(l, m)_A$ [$(l, m)_B$] locates at the middle point of $(l, m)_C$ and $(l+1, m)_C$ [$(l, m)_C$ and $(l, m+1)_C$]. The orange dot lines represent the couplings between the two sublattices. The butterfly-like spectra are shown in (b) for $\lambda = 0.9$, (c) for $\lambda = 1$, and (d) for $\lambda = 1.1$. The other parameters are chosen as $V_x/J_x = 10$, $k_x = 0$ and $L_y = 1000$.

Conclusion remarks. – In summary, from the collectively translational invariance, we introduce an intrinsic topological invariant for IQS's. Our topological invariant generalizes the TKNN invariant. As an application, we use our topological invariant to study the two-magnon excitations in a strongly interacting spinor Hofstadter

model with one particle per site. We explore the non-trivial topology of these excitations and demonstrate the emergence of topological edge BS's. We further give the topological phase diagram for the lowest BS subband. To understand the topological BS's, we derive an effective single-particle model described by a Hofstadter superlattice with two coupled standard Hofstadter sublattices. Our results provides a new way of characterizing correlated topological states in strongly IQS's.

At last, we briefly discuss the experimental possibility. Using the laser-assisted tunneling of Bose atoms in a tilted optical lattice, the 2D spinor Hofstadter model can be realized [10, 11] (see the Supplementary Material [24]). In addition, the interaction strength can be tuned via Feshbach resonances [30, 31]. Under strong interactions, the magnon excitations in the interacting spinor Hofstadter model obeys a 2D hard-core Bose-Hubbard model subjected to gauge fields. The selective magnon excitations can be prepared by using a line-shaped laser beam generated with a spatial light modulator [32, 33]. The two-magnon bound states can be observed by using the *in situ* correlation measurement [33].

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- [24] In our Supplementary Material, we give more details about: (i) the realization of the interacting spinor Hofstadter model, (ii) the derivation of the effective hard-core Bose-Hubbard model, (iii) the cotranslational symmetry of two-magnon excitations, and (iv) the derivation of the effective single-particle model for two-magnon bound-states.
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Supplementary Material for “An Intrinsic Topological Invariant in Strongly Interacting Quantum Systems”

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I. REALIZATION OF THE INTERACTING SPINOR HOFSTADTER MODEL

In this section, we give a detailed derivation of the interacting spinor Hofstadter model. Based upon the approach for treating noninteracting spinless bosons [S1], we generalize it to deal with interacting two-component bosons.

We consider an ultracold two-component Bose gas confined in a two-dimensional optical lattice potential,

$$V_{\text{latt}}(\mathbf{r}) = \frac{V_{x0}}{2} \cos\left(\frac{2\pi}{d_x}x\right) + \frac{V_{y0}}{2} \cos\left(\frac{2\pi}{d_y}y\right), \quad (\text{S.1})$$

with $\mathbf{r} = (x, y)$ and $d_\alpha = \lambda_\alpha/2$. Here, λ_α and $V_{\alpha 0}$ are respectively the wavelength and lattice depth along α -direction (where $\alpha = x$ and y). A gradient magnetic field along x -direction is used to generate a spin-dependent linear potential,

$$V_{\text{til}}(\mathbf{r}) = \frac{\Delta}{d_x} x \hat{\sigma}_z, \quad (\text{S.2})$$

with the amplitude Δ . Given the bare coupling along x -direction t_x , when $\Delta \gg t_x$, the tunneling along x -direction is inhibited and can be restored by a pair of far-detuned running-wave beams,

$$V_K(\mathbf{r}, t) = \Omega \cos(\mathbf{k}' \cdot \mathbf{r} - \omega t), \quad (\text{S.3})$$

with $\omega = \omega_1 - \omega_2 = \Delta/\hbar$ and $\mathbf{k}' = \mathbf{k}_1 - \mathbf{k}_2 = (k'_x, k'_y)$, see Fig. S1.

If the atom-atom interactions are dominated by two-body interactions, the many-body Hamiltonian includes two parts: a one-body part for single-particle contributions and a two-body part for atom-atom interactions. The single-particle Hamiltonian reads,

$$\hat{h}_0 = \frac{\hat{\mathbf{p}}^2}{2M} + V_{\text{latt}}(\mathbf{r}) + V_{\text{til}}(\mathbf{r}) + V_K(\mathbf{r}, t). \quad (\text{S.4})$$

Here, $\hat{\mathbf{p}} = (\hat{p}_x, \hat{p}_y)$ and M is the atomic mass. Under ultralow temperature, the atom-atom interaction is

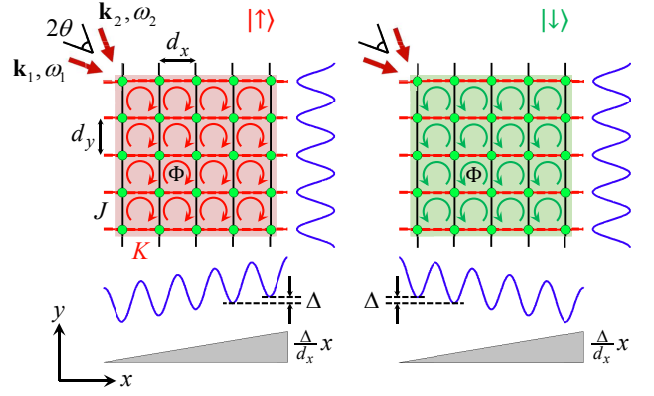


FIG. S1. (color online). Schematic diagram. The ultracold Bose atoms are confined in a two-dimensional optical lattice. The lattice constants are given as $d_\alpha = \lambda_\alpha/2$ with $\alpha = x$ and y . Along y -direction, the nearest-neighboring tunneling occurs with strength t_y . Along x -direction, the nearest-neighboring tunneling is affected by a magnetic field gradient Δ/d_x , which introduces an energy offset between neighboring sites of (left) Δ for $|\uparrow\rangle$ atoms and (right) $-\Delta$ for $|\downarrow\rangle$ atoms. An additional pair of laser beams with wave vectors $|\mathbf{k}_1| \simeq |\mathbf{k}_2| = 2\pi/\lambda_K$ and frequency difference $\omega = \omega_1 - \omega_2$ is used to restore resonant tunneling with complex amplitude K . This realizes an effective flux of $\Phi = k'_y d_y$ [where $\mathbf{k}' = \mathbf{k}_1 - \mathbf{k}_2 = (k'_x, k'_y)$] for $|\uparrow\rangle$ bosons (left) and $-\Phi$ for $|\downarrow\rangle$ bosons (right).

described by the s -wave scattering and the many-body Hamiltonian reads,

$$\begin{aligned} \hat{H} = & \int d^2\mathbf{r} \left[\hat{\psi}^\dagger(\mathbf{r}) \hat{h}_0 \hat{\psi}(\mathbf{r}) \right] \\ & + \sum_{\sigma_1, \sigma_2} \frac{1}{2} g_{\sigma_1 \sigma_2} \int d^2\mathbf{r} \left[\hat{\psi}_{\sigma_1}^\dagger(\mathbf{r}) \hat{\psi}_{\sigma_2}^\dagger(\mathbf{r}) \hat{\psi}_{\sigma_2}(\mathbf{r}) \hat{\psi}_{\sigma_1}(\mathbf{r}) \right], \end{aligned} \quad (\text{S.5})$$

with the field operators $\hat{\psi}^\dagger(\mathbf{r}) = [\hat{\psi}_\uparrow^\dagger(\mathbf{r}), \hat{\psi}_\downarrow^\dagger(\mathbf{r})]$, which creates a boson at position \mathbf{r} with $|\uparrow\rangle, |\downarrow\rangle$. The interaction strength is given as $g_{\sigma_1 \sigma_2} = \frac{4\pi\hbar^2}{M} a_{\sigma_1 \sigma_2}$ with $a_{\sigma_1 \sigma_2}$ denot-

ing the s -wave scattering length between components σ_1 and σ_2 . Introducing $\gamma_\uparrow = 1$ and $\gamma_\downarrow = -1$, the many-body Hamiltonian becomes,

$$\hat{H} = \sum_{\sigma} \int d^2\mathbf{r} \left[\hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) \hat{h}_{0,\sigma} \hat{\psi}_{\sigma}(\mathbf{r}) \right] + \sum_{\sigma_1, \sigma_2} \frac{1}{2} g_{\sigma_1 \sigma_2} \int d^2\mathbf{r} \left[\hat{\psi}_{\sigma_1}^{\dagger}(\mathbf{r}) \hat{\psi}_{\sigma_2}^{\dagger}(\mathbf{r}) \hat{\psi}_{\sigma_2}(\mathbf{r}) \hat{\psi}_{\sigma_1}(\mathbf{r}) \right] \quad (\text{S.6})$$

with

$$\hat{h}_{0,\sigma} = \frac{\hat{\mathbf{p}}^2}{2M} + V_{\text{latt}}(\mathbf{r}) + \gamma_{\sigma} \frac{\Delta}{d_x} x + V_K(\mathbf{r}, t). \quad (\text{S.7})$$

Although the system may involve multiple bands, we assume our system only involves the lowest band which can be realized when the optical lattice is sufficiently deep.

Now we consider the Wannier-Stark-Wannier (WS-W) functions for the lowest band,

$$\phi_{\sigma}(\mathbf{r} - \mathbf{r}_{l,m}) = \phi_{\sigma}^{\text{ws}x}(x - x_l) \phi^{\text{wy}}(y - y_m), \quad (\text{S.8})$$

with $\mathbf{r}_{l,m} = (x_l, y_m) = (ld_x, md_y)$. Define $\hat{h}_{\alpha} = \frac{\hat{p}_{\alpha}^2}{2M} + \frac{V_{\alpha 0}}{2} \cos(\frac{2\pi}{d_{\alpha}} \alpha)$ for $\alpha = x$ and y , we have $\phi_{\sigma}^{\text{ws}x}(x - x_l)$ being the Wannier-Stark function for $\hat{h}_{x,\sigma} = \hat{h}_x + \alpha_{\sigma} \frac{\Delta}{d_x} x$, while $\phi^{\text{wy}}(y - y_m)$ being the Wannier function for \hat{h}_y . By using the Wannier functions $\phi^{\text{ws}x}(x - x_j)$ for \hat{h}_x , the Wannier-Stark functions $\phi_{\sigma}^{\text{ws}x}(x - x_l)$ can be expanded as,

$$\phi_{\sigma}^{\text{ws}x}(x - x_l) = \sum_j J_{l-j}(\gamma_{\sigma}) \phi^{\text{ws}x}(x - x_j), \quad (\text{S.9})$$

with $\gamma_{\sigma} = \alpha_{\sigma} 2t_x / \Delta$ and $J_{\nu}(z)$ being the ν -order Bessel function of the first kind. Where, the bare tunnelling strengths along x - and y -directions are denoted as t_x and t_y respectively.

By using the WS-W basis, the field operators can be expanded as,

$$\hat{\psi}_{\sigma}^{\dagger}(\mathbf{r}) = \sum_{l,m} \phi_{\sigma}^*(\mathbf{r} - \mathbf{r}_{l,m}) \hat{a}_{l,m,\sigma}^{\dagger}, \quad (\text{S.10})$$

where $\hat{a}_{l,m,\sigma}^{\dagger}$ creates a σ -component boson at the (l, m) -th lattice site. Thus the many-body Hamiltonian reads,

$$\begin{aligned} \hat{H} = & \sum_{l',m',l,m,\sigma} t_{l',m';l,m}^{\sigma} \hat{a}_{l',m',\sigma}^{\dagger} \hat{a}_{l,m,\sigma} + \sum_{l',m',l,m,\sigma} V_{l',m';l,m}^{\sigma}(t) \hat{a}_{l',m',\sigma}^{\dagger} \hat{a}_{l,m,\sigma} \\ & + \sum_{l',m',l'_2,m'_2;l_2,m_2,l,m,\sigma_1,\sigma_2} U_{l',m';l'_2,m'_2;l_2,m_2,l,m}^{\sigma_1,\sigma_2} \hat{a}_{l',m',\sigma_1}^{\dagger} \hat{a}_{l'_2,m'_2,\sigma_2}^{\dagger} \hat{a}_{l_2,m_2,\sigma_2} \hat{a}_{l,m,\sigma_1}, \end{aligned} \quad (\text{S.11})$$

with the parameters

$$\begin{cases} t_{l',m';l,m}^{\sigma} = \int d^2\mathbf{r} \phi_{\sigma}^*(\mathbf{r} - \mathbf{r}_{l',m'}) (\hat{h}_{x,\sigma} + \hat{h}_y) \phi_{\sigma}(\mathbf{r} - \mathbf{r}_{l,m}) \\ V_{l',m';l,m}^{\sigma}(t) = \int d^2\mathbf{r} \phi_{\sigma}^*(\mathbf{r} - \mathbf{r}_{l',m'}) V_K(\mathbf{r}, t) \phi_{\sigma}(\mathbf{r} - \mathbf{r}_{l,m}) \\ U_{l',m';l'_2,m'_2;l_2,m_2,l,m}^{\sigma_1,\sigma_2} = \frac{1}{2} g_{\sigma_1 \sigma_2} \int d^2\mathbf{r} \phi_{\sigma_1}^*(\mathbf{r} - \mathbf{r}_{l',m'}) \phi_{\sigma_2}^*(\mathbf{r} - \mathbf{r}_{l'_2,m'_2}) \phi_{\sigma_2}(\mathbf{r} - \mathbf{r}_{l_2,m_2}) \phi_{\sigma_1}(\mathbf{r} - \mathbf{r}_{l,m}). \end{cases} \quad (\text{S.12})$$

Under the single-band tight-binding (SBTB) approximation, we have

$$t_{l',m';l,m}^{\sigma} = \alpha_{\sigma} \Delta l \delta_l^{\prime\prime} \delta_m^{m'} + t^y \delta_l^{\prime\prime} (\delta_{m+1}^{m'} + \delta_{m-1}^{m'}), \quad (\text{S.13})$$

$$U_{l',m';l'_2,m'_2;l_2,m_2,l,m}^{\sigma_1,\sigma_2} = \frac{1}{2} U_{\sigma_1 \sigma_2} \delta_l^{\prime\prime} \delta_m^{m'} \delta_{l'_2}^{l'_2} \delta_{m'_2}^{m'_2} \delta_{l_2}^{l_2} \delta_{m_2}^{m_2}, \quad (\text{S.14})$$

with

$$U_{\sigma_1 \sigma_2} = g_{\sigma_1 \sigma_2} \int dx |\phi_{\sigma_1}^{\text{ws}x}(x) \phi_{\sigma_2}^{\text{ws}x}(x)|^2 \int dy |\phi^{\text{wy}}(y)|^4. \quad (\text{S.15})$$

The matrix elements of $V_K(\mathbf{r}, t)$ are given as,

$$\begin{aligned} V_{l',m';l,m}^{\sigma}(t) = & \Omega \int dx \int dy [\phi_{\sigma}^{\text{ws}x*}(x) \phi_{\sigma}^{\text{ws}x}(x - x_{l-l'}) \\ & \times \phi^{\text{wy}*}(y) \phi^{\text{wy}}(y - y_{m-m'}) \\ & \times \cos(k'_x x + k'_y y - \theta_{l',m'})], \end{aligned} \quad (\text{S.16})$$

with $\theta_{l',m'} = \omega t - \phi_{l',m'}$, and $\phi_{l',m'} = l' \phi_x + m' \phi_y$ with

$\phi_x = k'_x d_x$ and $\phi_y = k'_y d_y$. Define

$$\begin{cases} I_{\sigma,l-l'}^{x,\cos} = \int dx \phi_{\sigma}^{\text{ws}x*}(x) \phi_{\sigma}^{\text{ws}x}(x - x_{l-l'}) \cos(k'_x x) \\ I_{\sigma,l-l'}^{x,\sin} = \int dx \phi_{\sigma}^{\text{ws}x*}(x) \phi_{\sigma}^{\text{ws}x}(x - x_{l-l'}) \sin(k'_x x) \\ I_{m-m'}^{y,\cos} = \int dy \phi^{\text{ws}y*}(y) \phi^{\text{ws}y}(y - y_{m-m'}) \cos(k'_y y) \\ I_{m-m'}^{y,\sin} = \int dy \phi^{\text{ws}y*}(y) \phi^{\text{ws}y}(y - y_{m-m'}) \sin(k'_y y) \end{cases}, \quad (\text{S.17})$$

as $\cos(k'_x x + k'_y y - \theta_{l',m'}) = \cos(k'_x x) \cos(k'_y y) \cos(\theta_{l',m'}) + \cos(k'_x x) \sin(k'_y y) \sin(\theta_{l',m'}) + \sin(k'_x x) \cos(k'_y y) \sin(\theta_{l',m'}) - \sin(k'_x x) \sin(k'_y y) \cos(\theta_{l',m'})$, we have

$$\begin{cases} I_{\sigma,l-l'}^{x,\cos} = \delta_l^{l'} I_{\sigma,0}^{x,\cos} + \delta_{l-1}^{l'} I_{\sigma,1}^{x,\cos} + \delta_{l+1}^{l'} I_{\sigma,-1}^{x,\cos} \\ I_{\sigma,l-l'}^{x,\sin} = \delta_l^{l'} I_{\sigma,0}^{x,\sin} + \delta_{l-1}^{l'} I_{\sigma,1}^{x,\sin} + \delta_{l+1}^{l'} I_{\sigma,-1}^{x,\sin} \\ I_{m-m'}^{y,\cos} = \delta_m^{m'} I_0^{y,\cos} \\ I_{m-m'}^{y,\sin} = \delta_m^{m'} I_0^{y,\sin} \end{cases}. \quad (\text{S.18})$$

under the SBTB approximation.

There are several different types of Wannier functions, it is better to use the maximally localized Wannier functions for constructing $\phi^{\text{ws}x}(x - x_j)$ and $\phi^{\text{ws}y}(y - y_m)$. The symmetry of the lattice potential implies the symmetric nature of the maximally localized Wannier functions [S2] (i.e., they are either symmetric or antisymmetric). Therefore, under the SBTB approximation, we have the following identities: $I_0^{y,\sin} = 0$, $I_{\sigma,0}^{x,\sin} = 0$, and $I_{\sigma,1}^{x,\cos} \cos(\theta_{l,m}) + I_{\sigma,1}^{x,\sin} \sin(\theta_{l,m}) = I_{\sigma,-1}^{x,\cos} \cos(\theta_{l+1,m}) + I_{\sigma,-1}^{x,\sin} \sin(\theta_{l+1,m})$. As $I_{\sigma,0}^{x,\cos}$ is σ -independent, one can define $I_0^x = I_{\sigma,0}^{x,\cos}$ and $I_0^y = I_0^{y,\cos}$, therefore one can obtain

$$\begin{aligned} V_{l',m';l,m}^{\sigma} &= \Omega I_0^y \delta_m^{m'} \{ \delta_l^{l'} I_0^x \cos(\theta_{l,m}) \\ &\quad + \delta_{l-1}^{l'} [I_{\sigma,1}^{x,\cos} \cos(\theta_{l-1,m}) + I_{\sigma,1}^{x,\sin} \sin(\theta_{l-1,m})] \\ &\quad + \delta_{l+1}^{l'} [I_{\sigma,-1}^{x,\cos} \cos(\theta_{l+1,m}) + I_{\sigma,-1}^{x,\sin} \sin(\theta_{l+1,m})] \}. \end{aligned} \quad (\text{S.19})$$

From Eqs. (S.11), (S.13), (S.14), and (S.19), the SBTB Hamiltonian can be written as,

$$\hat{H} = \hat{H}^D + \hat{H}^{\text{OD}x} + \hat{H}^{\text{OD}y} + \hat{H}^{\text{DI}}, \quad (\text{S.20})$$

with

$$\hat{H}^D = \sum_{l,m,\sigma} [\alpha_{\sigma} \Delta l + \Omega I_0^x I_0^y \cos(\theta_{l,m})] \hat{n}_{l,m,\sigma}, \quad (\text{S.21})$$

$$\hat{H}^{\text{OD}x} = \sum_{l,m,\sigma} \Omega I_0^y I_{l,m,\sigma}^x \left(\hat{a}_{l+1,m,\sigma}^{\dagger} \hat{a}_{l,m,\sigma} + \text{h.c.} \right), \quad (\text{S.22})$$

$$\hat{H}^{\text{OD}y} = \sum_{l,m,\sigma} t^y \left(\hat{a}_{l,m+1,\sigma}^{\dagger} \hat{a}_{l,m,\sigma} + \text{h.c.} \right), \quad (\text{S.23})$$

$$\hat{H}^{\text{DI}} = \sum_{l,m,\sigma_1,\sigma_2} \frac{1}{2} U_{\sigma_1\sigma_2} \hat{n}_{l,m,\sigma_1} (\hat{n}_{l,m,\sigma_2} - \delta_{\sigma_2}^{\sigma_1}), \quad (\text{S.24})$$

where $\hat{n}_{l,m,\sigma} = \hat{a}_{l,m,\sigma}^{\dagger} \hat{a}_{l,m,\sigma}$ and $I_{l,m,\sigma}^x = I_{\sigma,1}^{x,\cos} \cos(\theta_{l,m}) + I_{\sigma,1}^{x,\sin} \sin(\theta_{l,m})$.

The time dependence of the diagonal term \hat{H}^D can be eliminated via a unitary transformation,

$$\hat{U} = \exp \left(i \sum_{l,m,\sigma} \Lambda_{l,m,\sigma} \hat{n}_{l,m,\sigma} \right), \quad (\text{S.25})$$

where,

$$\Lambda_{l,m,\sigma} = -\alpha_{\sigma} l \omega t - \frac{\Omega}{\hbar \omega} I_0^x I_0^y \sin(\theta_{l,m}) + l \theta_{\sigma} \quad (\text{S.26})$$

is real and time-dependent. For convenience, we introduce a spin-dependent phase θ_{σ} whose value will be determined below. The Hamiltonian in the rotating frame is given as $\hat{H}' = \hat{U}^{\dagger} \hat{H} \hat{U} - i \hbar \hat{U}^{\dagger} (\partial_t \hat{U})$. For a resonant driving (i.e. $\hbar \omega = \Delta$), we have $\hat{U}^{\dagger} \hat{H}^D \hat{U} - i \hbar \hat{U}^{\dagger} (\partial_t \hat{U}) = 0$. Thus, \hat{H}' becomes as

$$\hat{H}' = \hat{U}^{\dagger} \hat{H}^{\text{OD}x} \hat{U} + \hat{U}^{\dagger} \hat{H}^{\text{OD}y} \hat{U} + \hat{H}^{\text{DI}}. \quad (\text{S.27})$$

Using the bosonic identity $e^{-i\theta \hat{n}} \hat{a}^{\dagger} e^{i\theta \hat{n}} = e^{-i\theta} \hat{a}^{\dagger}$, we have $\hat{U}^{\dagger} \hat{a}_{l,m,\sigma}^{\dagger} \hat{U} = e^{-i\Lambda_{l,m,\sigma}} \hat{a}_{l,m,\sigma}^{\dagger}$ and $\hat{U}^{\dagger} \hat{a}_{l,m,\sigma} \hat{U} = e^{i\Lambda_{l,m,\sigma}} \hat{a}_{l,m,\sigma}$. Consequently, one can find

$$\hat{U}^{\dagger} \hat{a}_{l+1,m,\sigma}^{\dagger} \hat{a}_{l,m,\sigma} \hat{U} = e^{i(\Lambda_{l,m,\sigma} - \Lambda_{l+1,m,\sigma})} \hat{a}_{l+1,m,\sigma}^{\dagger} \hat{a}_{l,m,\sigma}, \quad (\text{S.28})$$

$$\hat{U}^{\dagger} \hat{a}_{l,m+1,\sigma}^{\dagger} \hat{a}_{l,m,\sigma} \hat{U} = e^{i(\Lambda_{l,m,\sigma} - \Lambda_{l,m+1,\sigma})} \hat{a}_{l,m+1,\sigma}^{\dagger} \hat{a}_{l,m,\sigma}, \quad (\text{S.29})$$

with the time-dependent phases

$$\Lambda_{l,m,\sigma} - \Lambda_{l+1,m,\sigma} = \alpha_{\sigma} \omega t - \theta_{\sigma} - \Gamma_x \cos(\omega t - \phi_{l,m} - \frac{\phi_x}{2}), \quad (\text{S.30})$$

$$\Lambda_{l,m,\sigma} - \Lambda_{l,m+1,\sigma} = -\Gamma_y \cos(\omega t - \phi_{l,m} - \frac{\phi_y}{2}). \quad (\text{S.31})$$

Here, $\Gamma_{\alpha} = \frac{2\Omega}{\hbar \omega} I_0^x I_0^y \sin(\frac{1}{2} \phi_{\alpha})$ with $\alpha = x$ and y . Using the variant of the Jacobi-Anger identity, $e^{-iz \cos(\theta)} = e^{iz \sin(\theta - \frac{\pi}{2})} = \sum_r J_r(z) e^{ir(\theta - \frac{\pi}{2})}$, the phase factors are given as

$$\begin{aligned} e^{i(\Lambda_{l,m,\sigma} - \Lambda_{l+1,m,\sigma})} &= \sum_r [J_r(\Gamma_x) e^{i(\alpha_{\sigma} + r)\omega t} \\ &\quad \times e^{-ir(\phi_{l,m} + \frac{\phi_x}{2} + \frac{\pi}{2}) - i\theta_{\sigma}}], \end{aligned} \quad (\text{S.32})$$

$$e^{i(\Lambda_{l,m,\sigma} - \Lambda_{l,m+1,\sigma})} = \sum_r J_r(\Gamma_y) e^{ir\omega t - ir(\phi_{l,m} + \frac{\phi_y}{2} + \frac{\pi}{2})}. \quad (\text{S.33})$$

Therefore the off-diagonal terms of the Hamiltonian \hat{H}' become as

$$\hat{U}^{\dagger} \hat{H}^{\text{OD}x} \hat{U} = \sum_{l,m,\sigma} [K_{l,m}^{\sigma}(t) \hat{a}_{l+1,m,\sigma}^{\dagger} \hat{a}_{l,m,\sigma} + \text{h.c.}] \quad (\text{S.34})$$

$$\hat{U}^{\dagger} \hat{H}^{\text{OD}y} \hat{U} = \sum_{l,m,\sigma} [J_{l,m}(t) \hat{a}_{l,m+1,\sigma}^{\dagger} \hat{a}_{l,m,\sigma} + \text{h.c.}] \quad (\text{S.35})$$

with

$$\begin{aligned} K_{l,m}^{\sigma}(t) &= \Omega I_0^y I_{l,m,\sigma}^x \sum_r [J_r(\Gamma_x) e^{i(\alpha_{\sigma} + r)\omega t} \\ &\quad \times e^{-ir(\phi_{l,m} + \frac{\phi_x}{2} + \frac{\pi}{2}) - i\theta_{\sigma}}] \end{aligned} \quad (\text{S.36})$$

$$J_{l,m}(t) = t^y \sum_r J_r(\Gamma_y) e^{ir\omega t - ir(\phi_{l,m} + \frac{\phi_y}{2} + \frac{\pi}{2})}. \quad (\text{S.37})$$

Time-averaging over a period of $2\pi/\omega$ and using the identity $\frac{1}{2\pi/\omega} \int_0^{2\pi/\omega} dt e^{ir\omega t} = \delta_{r,0}$ (for any integer r), one can obtain

$$\begin{cases} \frac{1}{2\pi/\omega} \int_0^{2\pi/\omega} dt K_{l,m}^\sigma(t) = e^{i\alpha_\sigma \phi_{l,m}} \tilde{K}^\sigma, \\ \frac{1}{2\pi/\omega} \int_0^{2\pi/\omega} dt J_{l,m}(t) = J. \end{cases} \quad (\text{S.38})$$

Here, $J = t^y J_0(\Gamma_y)$ and the σ -dependent constant \tilde{K}^σ is given as

$$\begin{aligned} \tilde{K}^\sigma &= \frac{1}{2} \Omega I_0^y e^{-i\theta_\sigma} [(I_{\sigma,1}^{x,\text{exp}})^* J_{1+\alpha_\sigma}(\Gamma_x) e^{i\frac{1}{2}(\alpha_\sigma+1)(\phi_x+\pi)} \\ &\quad + I_{\sigma,1}^{x,\text{exp}} J_{1-\alpha_\sigma}(\Gamma_x) e^{i\frac{1}{2}(\alpha_\sigma-1)(\phi_x+\pi)}] \end{aligned} \quad (\text{S.39})$$

with the notation $I_{\sigma,1}^{x,\text{exp}} = I_{\sigma,1}^{x,\text{cos}} + iI_{\sigma,1}^{x,\text{sin}}$. Notice that $(I_{\sigma,1}^{x,\text{exp}})^* = e^{-i\phi_x} I_{\sigma,1}^{x,\text{exp}}$, if we define

$$\frac{1}{2} \Omega I_0^y [I_{\uparrow,1}^{x,\text{exp}} J_0(\Gamma_x) - I_{\downarrow,1}^{x,\text{exp}} J_2(\Gamma_x)] \equiv K e^{i\theta_K} \quad (\text{S.40})$$

with $K > 0$ and $\theta_K \in (-\pi, \pi]$, we have

$$\begin{cases} \tilde{K}^\uparrow = K e^{i(-\theta_\uparrow + \theta_K)} \\ \tilde{K}^\downarrow = K e^{-i(\theta_\downarrow + \phi_x - \theta_K)} \end{cases} \quad (\text{S.41})$$

The undetermined phases θ_σ are thus given as $\theta_\uparrow = \theta_K$ and $\theta_\downarrow = \theta_K - \phi_x$ such that $\tilde{K}^\uparrow = \tilde{K}^\downarrow = K > 0$. Thus the effective Hamiltonian in the rotating frame $\hat{H}_{\text{eff}} = \frac{1}{2\pi/\omega} \int_0^{2\pi/\omega} dt \hat{H}'$ is given as

$$\begin{aligned} \hat{H}_{\text{eff}} &= \sum_{l,m,\sigma} \left(K e^{i\alpha_\sigma \phi_{l,m}} \hat{a}_{l+1,m,\sigma}^\dagger \hat{a}_{l,m,\sigma} + \text{h.c.} \right) \\ &\quad + \sum_{l,m,\sigma} \left(J \hat{a}_{l,m+1,\sigma}^\dagger \hat{a}_{l,m,\sigma} + \text{h.c.} \right) \\ &\quad + \sum_{l,m,\sigma_1,\sigma_2} \frac{1}{2} U_{\sigma_1\sigma_2} \hat{n}_{l,m,\sigma_1} (\hat{n}_{l,m,\sigma_2} - \delta_{\sigma_2}^{\sigma_1}). \end{aligned} \quad (\text{S.42})$$

Through a time-independent unitary transformation,

$$\hat{U}' = \exp \left(i \sum_{l,m,\sigma} \Lambda'_{l,m,\sigma} \hat{n}_{l,m,\sigma} \right), \quad (\text{S.43})$$

where $\Lambda'_{l,m,\sigma} = \alpha_\sigma [\frac{1}{2}\phi_x l^2 - (\pi + \frac{1}{2}\phi_x)l] - m\pi$, one can change the sign of K and J . Since $\hat{U}'^\dagger \hat{a}_{l,m,\sigma}^\dagger \hat{U}' = e^{-i\Lambda'_{l,m,\sigma}} \hat{a}_{l,m,\sigma}^\dagger$ and $\hat{U}'^\dagger \hat{a}_{l,m,\sigma} \hat{U}' = e^{i\Lambda'_{l,m,\sigma}} \hat{a}_{l,m,\sigma}$, we have

$$\hat{U}'^\dagger \hat{a}_{l+1,m,\sigma}^\dagger \hat{a}_{l,m,\sigma} \hat{U}' = e^{i(\Lambda'_{l,m,\sigma} - \Lambda'_{l+1,m,\sigma})} \hat{a}_{l+1,m,\sigma}^\dagger \hat{a}_{l,m,\sigma}, \quad (\text{S.44})$$

$$\hat{U}'^\dagger \hat{a}_{l,m+1,\sigma}^\dagger \hat{a}_{l,m,\sigma} \hat{U}' = e^{i(\Lambda'_{l,m,\sigma} - \Lambda'_{l,m+1,\sigma})} \hat{a}_{l,m+1,\sigma}^\dagger \hat{a}_{l,m,\sigma}, \quad (\text{S.45})$$

with the phases

$$\Lambda'_{l,m,\sigma} - \Lambda'_{l+1,m,\sigma} = \alpha_\sigma (\pi - l\phi_x), \quad (\text{S.46})$$

$$\Lambda'_{l,m,\sigma} - \Lambda'_{l,m+1,\sigma} = \pi. \quad (\text{S.47})$$

Thus the effective Hamiltonian becomes

$$\begin{aligned} \hat{H}_B &= - \sum_{l,m,\sigma} \left[K e^{i\alpha_\sigma m \Phi} \hat{a}_{l+1,m,\sigma}^\dagger \hat{a}_{l,m,\sigma} + \text{h.c.} \right] \\ &\quad - \sum_{l,m,\sigma} \left[J \hat{a}_{l,m+1,\sigma}^\dagger \hat{a}_{l,m,\sigma} + \text{h.c.} \right] \\ &\quad + \sum_{l,m,\sigma_1,\sigma_2} \frac{1}{2} U_{\sigma_1\sigma_2} \hat{n}_{l,m,\sigma_1} (\hat{n}_{l,m,\sigma_2} - \delta_{\sigma_2}^{\sigma_1}) \end{aligned} \quad (\text{S.48})$$

with $\Phi = \phi_y = k'_y d_y$. The above Hamiltonian is an interacting spinor Hofstadter model.

II. DERIVATION OF THE EFFECTIVE HARD-CORE BOSE-HUBBARD MODEL

In the strongly interacting regime [any of $(U_{\uparrow\uparrow}, U_{\uparrow\downarrow}, U_{\downarrow\downarrow})$ is far larger than any of (K, J)], the model (S.48) with unity filling can be mapped onto a spin model, which is equivalent to a hard-core Bose-Hubbard model. Below, by using the perturbation theory for degenerated many-body quantum systems [S3], we analytically derive an effective spin model up to second-order perturbation.

In the strongly interacting regime, one can treat the hopping terms

$$\hat{H}_1 = \hat{H}_K + \hat{H}_J \quad (\text{S.49})$$

as a perturbation to the interaction term

$$\hat{H}_0 = \sum_{l,m,\sigma_1,\sigma_2} \frac{1}{2} U_{\sigma_1\sigma_2} \hat{n}_{l,m,\sigma_1} (\hat{n}_{l,m,\sigma_2} - \delta_{\sigma_2}^{\sigma_1}), \quad (\text{S.50})$$

where, the hopping terms are given as

$$\begin{cases} \hat{H}_K = -K \sum_{l,m} [\hat{T}_{l,m}^x + \hat{T}_{l,m}^{x\dagger}] \\ \hat{H}_J = -J \sum_{l,m} [\hat{T}_{l,m}^y + \hat{T}_{l,m}^{y\dagger}] \end{cases} \quad (\text{S.51})$$

with

$$\begin{cases} \hat{T}_{l,m}^x = \sum_{\sigma} e^{i\alpha_\sigma m \Phi} \hat{a}_{l+1,m,\sigma}^\dagger \hat{a}_{l,m,\sigma} \\ \hat{T}_{l,m}^y = \sum_{\sigma} \hat{a}_{l,m+1,\sigma}^\dagger \hat{a}_{l,m,\sigma} \end{cases} \quad (\text{S.52})$$

Obviously, any Fock state is an eigenstate of \hat{H}_0 . The Fock state for the system of unity filling is given as

$$|\mathbf{n}\rangle = |\dots, n_{l,m,\sigma}, \dots\rangle = \prod_{l,m,\sigma} \frac{1}{\sqrt{n_{l,m,\sigma}!}} (\hat{a}_{l,m,\sigma}^\dagger)^{n_{l,m,\sigma}} |\mathbf{0}\rangle \quad (\text{S.53})$$

where $\sum_{l,m,\sigma} n_{l,m,\sigma} = L_x L_y = L$ [L_α is the number of lattice sites along α -direction ($\alpha = x$ and y), while L is the total number of sites of the whole two-dimensional lattice] and $|\mathbf{0}\rangle$ denotes the vacuum state. According to the eigen-equation $\hat{H}_0 |\mathbf{n}\rangle = E_{\mathbf{n}} |\mathbf{n}\rangle$, we have the eigen-energy,

$$E_{\mathbf{n}} = \sum_{l,m,\sigma_1,\sigma_2} \frac{1}{2} U_{\sigma_1\sigma_2} n_{l,m,\sigma_1} (n_{l,m,\sigma_2} - \delta_{\sigma_2}^{\sigma_1}). \quad (\text{S.54})$$

Obviously, due to only one atom in each lattice site, the ground-state has energy $E_0 = 0$ and 2^L -fold degeneracy. In the Fock basis, the ground states are expressed as

$$|\mathbf{s}\rangle = |\dots, s_{l,m}, \dots\rangle = \prod_{l,m} \hat{a}_{l,m,\sigma_{lm}}^\dagger |\mathbf{0}\rangle, \quad (\text{S.55})$$

with $s_{l,m} \in \{\uparrow, \downarrow\}$ and $\hat{H}_0 |\mathbf{s}\rangle = E_0 |\mathbf{s}\rangle$.

The projector onto the ground-state space \mathcal{U}_0 is,

$$\hat{P}_0 = \sum_{\mathbf{s}} |\mathbf{s}\rangle \langle \mathbf{s}|. \quad (\text{S.56})$$

Introducing \mathcal{V}_0 as the orthogonal complement of \mathcal{U}_0 , the relevant projector onto \mathcal{V}_0 is,

$$\hat{S} = - \sum_{E_{\mathbf{n}} \neq 0} \frac{1}{E_{\mathbf{n}}} |\mathbf{n}\rangle \langle \mathbf{n}|. \quad (\text{S.57})$$

Thus the effective Hamiltonian up to 2nd order is given as,

$$\hat{H}_{\text{eff}}^{(2)} = \hat{P}_0 \hat{H}_1 \hat{S} \hat{H}_1 \hat{P}_0. \quad (\text{S.58})$$

It's easy to find that $\hat{P}_0 \hat{H}_K \hat{S} \hat{H}_J \hat{P}_0 = \hat{P}_0 \hat{H}_J \hat{S} \hat{H}_K \hat{P}_0 = 0$, which gives

$$\hat{H}_{\text{eff}}^{(2)} = \hat{P}_0 \hat{H}_K \hat{S} \hat{H}_K \hat{P}_0 + \hat{P}_0 \hat{H}_J \hat{S} \hat{H}_J \hat{P}_0. \quad (\text{S.59})$$

Furthermore, since

$$\begin{cases} \hat{P}_0 \hat{T}_{l,m}^\alpha \hat{S} \hat{T}_{l',m'}^\alpha \hat{P}_0 = \hat{P}_0 \hat{T}_{l,m}^{\alpha\dagger} \hat{S} \hat{T}_{l',m'}^{\alpha\dagger} \hat{P}_0 = 0 \\ \hat{P}_0 \hat{T}_{l,m}^\alpha \hat{S} \hat{T}_{l',m'}^{\alpha\dagger} \hat{P}_0 = \delta_l^{l'} \delta_m^{m'} \hat{P}_0 \hat{T}_{l,m}^\alpha \hat{S} \hat{T}_{l,m}^{\alpha\dagger} \hat{P}_0 \\ \hat{P}_0 \hat{T}_{l,m}^{\alpha\dagger} \hat{S} \hat{T}_{l',m'}^\alpha \hat{P}_0 = \delta_l^{l'} \delta_m^{m'} \hat{P}_0 \hat{T}_{l,m}^{\alpha\dagger} \hat{S} \hat{T}_{l,m}^\alpha \hat{P}_0 \end{cases} \quad (\text{S.60})$$

for $\alpha = x$ and y , we have

$$\hat{P}_0 \hat{H}_K \hat{S} \hat{H}_K \hat{P}_0 = K^2 \sum_{l,m} (\hat{P}_0 \hat{T}_{l,m}^x \hat{S} \hat{T}_{l,m}^{x\dagger} \hat{P}_0 + \hat{P}_0 \hat{T}_{l,m}^{x\dagger} \hat{S} \hat{T}_{l,m}^x \hat{P}_0), \quad (\text{S.61})$$

and

$$\hat{P}_0 \hat{H}_J \hat{S} \hat{H}_J \hat{P}_0 = J^2 \sum_{l,m} (\hat{P}_0 \hat{T}_{l,m}^y \hat{S} \hat{T}_{l,m}^{y\dagger} \hat{P}_0 + \hat{P}_0 \hat{T}_{l,m}^{y\dagger} \hat{S} \hat{T}_{l,m}^y \hat{P}_0). \quad (\text{S.62})$$

This means that the effective Hamiltonian has two parts, which respectively correspond to the influences from the hopping terms of x - and y -directions.

As the effective Hamiltonian only involves the nearest-neighbor couplings, it is sufficient to give its parameters by considering a system of two lattice sites. For the hopping along x -direction, we take site- (l, m) as site-1 and site- $(l+1, m)$ as site-2. The two-site ground-states are

$$\begin{cases} |\uparrow, \uparrow\rangle = \hat{a}_{1\uparrow}^\dagger \hat{a}_{2\uparrow}^\dagger |\mathbf{0}\rangle, & |\uparrow, \downarrow\rangle = \hat{a}_{1\uparrow}^\dagger \hat{a}_{2\downarrow}^\dagger |\mathbf{0}\rangle, \\ |\downarrow, \uparrow\rangle = \hat{a}_{1\downarrow}^\dagger \hat{a}_{2\uparrow}^\dagger |\mathbf{0}\rangle, & |\downarrow, \downarrow\rangle = \hat{a}_{1\downarrow}^\dagger \hat{a}_{2\downarrow}^\dagger |\mathbf{0}\rangle, \end{cases} \quad (\text{S.63})$$

with the eigenenergy $E_0 = 0$. While the two-site excited states are

$$\begin{cases} |\uparrow\uparrow, 0\rangle = \frac{1}{\sqrt{2}} (\hat{a}_{1\uparrow}^\dagger)^2 |\mathbf{0}\rangle, & |0, \uparrow\uparrow\rangle = \frac{1}{\sqrt{2}} (\hat{a}_{2\uparrow}^\dagger)^2 |\mathbf{0}\rangle, \\ |\downarrow\downarrow, 0\rangle = \frac{1}{\sqrt{2}} (\hat{a}_{1\downarrow}^\dagger)^2 |\mathbf{0}\rangle, & |0, \downarrow\downarrow\rangle = \frac{1}{\sqrt{2}} (\hat{a}_{2\downarrow}^\dagger)^2 |\mathbf{0}\rangle, \\ |\uparrow\downarrow, 0\rangle = \hat{a}_{1\uparrow}^\dagger \hat{a}_{1\downarrow}^\dagger |\mathbf{0}\rangle, & |0, \uparrow\downarrow\rangle = \hat{a}_{2\uparrow}^\dagger \hat{a}_{2\downarrow}^\dagger |\mathbf{0}\rangle, \end{cases} \quad (\text{S.64})$$

with eigenenergies $E_{\uparrow\uparrow,0} = E_{0,\uparrow\uparrow} = U_{\uparrow\uparrow}$, $E_{\downarrow\downarrow,0} = E_{0,\downarrow\downarrow} = U_{\downarrow\downarrow}$ and $E_{\uparrow\downarrow,0} = E_{0,\uparrow\downarrow} = U_{\uparrow\downarrow}$. Hence, we have the projectors

$$\begin{aligned} \hat{P}_0 = & [\hat{a}_{1\uparrow}^\dagger \hat{a}_{2\uparrow}^\dagger |\mathbf{0}\rangle \langle \mathbf{0}| \hat{a}_{2\uparrow} \hat{a}_{1\uparrow} + \hat{a}_{1\uparrow}^\dagger \hat{a}_{2\downarrow}^\dagger |\mathbf{0}\rangle \langle \mathbf{0}| \hat{a}_{2\downarrow} \hat{a}_{1\uparrow} \\ & + \hat{a}_{1\downarrow}^\dagger \hat{a}_{2\uparrow}^\dagger |\mathbf{0}\rangle \langle \mathbf{0}| \hat{a}_{2\uparrow} \hat{a}_{1\downarrow} + \hat{a}_{1\downarrow}^\dagger \hat{a}_{2\downarrow}^\dagger |\mathbf{0}\rangle \langle \mathbf{0}| \hat{a}_{2\downarrow} \hat{a}_{1\downarrow}], \end{aligned} \quad (\text{S.65})$$

$$\begin{aligned} \hat{S} = & - \left\{ \frac{1}{2U_{\uparrow\uparrow}} [(\hat{a}_{1\uparrow}^\dagger)^2 |\mathbf{0}\rangle \langle \mathbf{0}| (\hat{a}_{1\uparrow})^2 + (\hat{a}_{2\uparrow}^\dagger)^2 |\mathbf{0}\rangle \langle \mathbf{0}| (\hat{a}_{2\uparrow})^2] \right. \\ & + \frac{1}{2U_{\downarrow\downarrow}} [(\hat{a}_{1\downarrow}^\dagger)^2 |\mathbf{0}\rangle \langle \mathbf{0}| (\hat{a}_{1\downarrow})^2 + (\hat{a}_{2\downarrow}^\dagger)^2 |\mathbf{0}\rangle \langle \mathbf{0}| (\hat{a}_{2\downarrow})^2] \\ & \left. + \frac{1}{U_{\uparrow\downarrow}} [\hat{a}_{1\uparrow}^\dagger \hat{a}_{1\downarrow}^\dagger |\mathbf{0}\rangle \langle \mathbf{0}| \hat{a}_{1\downarrow} \hat{a}_{1\uparrow} + \hat{a}_{2\uparrow}^\dagger \hat{a}_{2\downarrow}^\dagger |\mathbf{0}\rangle \langle \mathbf{0}| \hat{a}_{2\downarrow} \hat{a}_{2\uparrow}] \right\}, \end{aligned} \quad (\text{S.66})$$

and

$$\hat{T}_{l,m}^x = e^{im\Phi} \hat{a}_{2\uparrow}^\dagger \hat{a}_{1\uparrow} + e^{-im\Phi} \hat{a}_{2\downarrow}^\dagger \hat{a}_{1\downarrow}. \quad (\text{S.67})$$

Inserting Eqs. (S.65), (S.66), and (S.67) into Eq. (S.61), and using the bosonic commutation relations and the identity $\hat{a}_{j,\sigma} \hat{a}_{j',\sigma'}^\dagger |\mathbf{0}\rangle = \delta_{j,j'}^\sigma \delta_{\sigma,\sigma'} |\mathbf{0}\rangle$ ($j, j' \in \{1, 2\}$), after some tedious algebra, we obtain

$$\begin{aligned} \hat{P}_0 \hat{T}_{l,m}^x \hat{S} \hat{T}_{l,m}^{x\dagger} \hat{P}_0 = & \hat{P}_0 \hat{T}_{l,m}^{x\dagger} \hat{S} \hat{T}_{l,m}^x \hat{P}_0 \\ = & - \left[\frac{2}{U_{\uparrow\uparrow}} \hat{a}_{1\uparrow}^\dagger \hat{a}_{2\uparrow}^\dagger \hat{a}_{2\uparrow} \hat{a}_{1\uparrow} + \frac{2}{U_{\downarrow\downarrow}} \hat{a}_{1\downarrow}^\dagger \hat{a}_{2\downarrow}^\dagger \hat{a}_{2\downarrow} \hat{a}_{1\downarrow} \right. \\ & + \frac{1}{U_{\uparrow\downarrow}} (\hat{a}_{1\downarrow}^\dagger \hat{a}_{2\uparrow}^\dagger \hat{a}_{2\uparrow} \hat{a}_{1\downarrow} + \hat{a}_{1\uparrow}^\dagger \hat{a}_{2\downarrow}^\dagger \hat{a}_{2\downarrow} \hat{a}_{1\uparrow}) \\ & \left. + \frac{1}{U_{\uparrow\downarrow}} (e^{i2m\Phi} \hat{a}_{1\downarrow}^\dagger \hat{a}_{2\uparrow}^\dagger \hat{a}_{2\downarrow} \hat{a}_{1\uparrow} + e^{-i2m\Phi} \hat{a}_{1\uparrow}^\dagger \hat{a}_{2\downarrow}^\dagger \hat{a}_{2\uparrow} \hat{a}_{1\downarrow}) \right]. \end{aligned} \quad (\text{S.68})$$

By introducing the pseudospin operators: $\hat{S}_{l,m}^+ = \hat{a}_{l,m,\uparrow}^\dagger \hat{a}_{l,m,\downarrow}$, $\hat{S}_{l,m}^- = \hat{a}_{l,m,\downarrow}^\dagger \hat{a}_{l,m,\uparrow}$, and $\hat{S}_{l,m}^z = \frac{1}{2}(\hat{n}_{l,m,\uparrow} - \hat{n}_{l,m,\downarrow})$ (we set $\hbar = 1$ here and after), Eq. (S.68) can be rewritten as

$$\begin{aligned} \hat{P}_0 \hat{T}_{l,m}^x \hat{S} \hat{T}_{l,m}^{x\dagger} \hat{P}_0 + \hat{P}_0 \hat{T}_{l,m}^{x\dagger} \hat{S} \hat{T}_{l,m}^x \hat{P}_0 = & - \left[2 \frac{1}{U_{\uparrow\downarrow}} (e^{i2m\Phi} \hat{S}_2^+ \hat{S}_1^- + e^{-i2m\Phi} \hat{S}_1^+ \hat{S}_2^-) \right. \\ & + 4 \left(\frac{1}{U_{\uparrow\uparrow}} + \frac{1}{U_{\downarrow\downarrow}} - \frac{1}{U_{\uparrow\downarrow}} \right) \hat{S}_1^z \hat{S}_2^z + 2 \left(\frac{1}{U_{\uparrow\uparrow}} - \frac{1}{U_{\downarrow\downarrow}} \right) (\hat{S}_1^z + \hat{S}_2^z) \\ & \left. + \left(\frac{1}{U_{\uparrow\uparrow}} + \frac{1}{U_{\downarrow\downarrow}} + \frac{1}{U_{\uparrow\downarrow}} \right) \right]. \end{aligned} \quad (\text{S.69})$$

Extended to the lattice, that is $1 \rightarrow (l, m)$ and $2 \rightarrow (l+1, m)$, we have

$$\begin{aligned} \hat{P}_0 \hat{T}_{l,m}^x \hat{S} \hat{T}_{l,m}^{x\dagger} \hat{P}_0 + \hat{P}_0 \hat{T}_{l,m}^{x\dagger} \hat{S} \hat{T}_{l,m}^x \hat{P}_0 = & - \left[2 \frac{1}{U_{\uparrow\downarrow}} (e^{i2m\Phi} \hat{S}_{l+1,m}^+ \hat{S}_{l,m}^- + e^{-i2m\Phi} \hat{S}_{l,m}^+ \hat{S}_{l+1,m}^-) \right. \\ & + 4 \left(\frac{1}{U_{\uparrow\uparrow}} + \frac{1}{U_{\downarrow\downarrow}} - \frac{1}{U_{\uparrow\downarrow}} \right) \hat{S}_{l,m}^z \hat{S}_{l+1,m}^z \\ & \left. + 2 \left(\frac{1}{U_{\uparrow\uparrow}} - \frac{1}{U_{\downarrow\downarrow}} \right) (\hat{S}_{l,m}^z + \hat{S}_{l+1,m}^z) + \left(\frac{1}{U_{\uparrow\uparrow}} + \frac{1}{U_{\downarrow\downarrow}} + \frac{1}{U_{\uparrow\downarrow}} \right) \right]. \end{aligned} \quad (\text{S.70})$$

For the hopping along y -direction, we take site- (l, m) as site-1 and site- $(l, m+1)$ as site-2. Similarly, up to the second-order perturbation, we obtain

$$\begin{aligned} & \hat{P}_0 \hat{T}_{l,m}^y \hat{S}_{l,m}^y \hat{T}_{l,m}^y \hat{P}_0 + \hat{P}_0 \hat{T}_{l,m}^y \hat{S}_{l,m}^y \hat{T}_{l,m}^y \hat{P}_0 \\ &= -[2\frac{1}{U_{\uparrow\downarrow}}(\hat{S}_{l,m+1}^+ \hat{S}_{l,m}^- + \hat{S}_{l,m}^+ \hat{S}_{l,m+1}^-) \\ &+ 4(\frac{1}{U_{\uparrow\uparrow}} + \frac{1}{U_{\downarrow\downarrow}} - \frac{1}{U_{\uparrow\downarrow}}) \hat{S}_{l,m}^z \hat{S}_{l,m+1}^z \\ &+ 2(\frac{1}{U_{\uparrow\uparrow}} - \frac{1}{U_{\downarrow\downarrow}})(\hat{S}_{l,m}^z \hat{S}_{l,m+1}^z) + (\frac{1}{U_{\uparrow\uparrow}} + \frac{1}{U_{\downarrow\downarrow}} + \frac{1}{U_{\uparrow\downarrow}})]. \end{aligned} \quad (S.71)$$

Introducing $J_x = 2K^2/U_{\uparrow\downarrow}$, $J_y = 2J^2/U_{\uparrow\downarrow}$, $V_x = 4K^2(1/U_{\uparrow\uparrow} + 1/U_{\downarrow\downarrow} - 1/U_{\uparrow\downarrow})$, $V_y = 4J^2(1/U_{\uparrow\uparrow} + 1/U_{\downarrow\downarrow} - 1/U_{\uparrow\downarrow})$, and $B_0 = 4(K^2 + J^2)(1/U_{\uparrow\uparrow} - 1/U_{\downarrow\downarrow})$, from Eqs. (S.59), (S.61), (S.62), (S.70), and (S.71), we get the effective Hamiltonian,

$$\begin{aligned} \hat{H}_{\text{eff}}^{(2)} = & - \sum_{l,m} [J_x e^{i2m\Phi} \hat{S}_{l+1,m}^+ \hat{S}_{l,m}^- + \text{h.c.}] \\ & - \sum_{l,m} [J_y \hat{S}_{l,m+1}^+ \hat{S}_{l,m}^- + \text{h.c.}] \\ & - \sum_{l,m} (V_x \hat{S}_{l,m}^z \hat{S}_{l+1,m}^z + V_y \hat{S}_{l,m}^z \hat{S}_{l,m+1}^z) \\ & - B_0 \sum_{l,m} \hat{S}_{l,m}^z. \end{aligned} \quad (S.72)$$

Here, we have removed a constant energy shift: $-(K^2 + J^2)(1/U_{\uparrow\uparrow} + 1/U_{\downarrow\downarrow} + 1/U_{\uparrow\downarrow})L_x L_y$.

According to the Matsubara-Matsuda mapping [S4]: $|\downarrow\rangle \leftrightarrow |0\rangle$, $|\uparrow\rangle \leftrightarrow |1\rangle$, $\hat{S}_{l,m}^+ \leftrightarrow \hat{b}_{l,m}^\dagger$, $\hat{S}_{l,m}^- \leftrightarrow \hat{b}_{l,m}$, and $\hat{S}_{l,m}^z \leftrightarrow (\hat{n}_{l,m} - \frac{1}{2}) \equiv (\hat{b}_{l,m}^\dagger \hat{b}_{l,m} - \frac{1}{2})$, the magnon excitations can be described by hard-core bosons and so that the two-dimensional Heisenberg spin model (S.72) is equivalent to a two-dimensional hard-core Bose-Hubbard model subjected to a synthetic gauge field,

$$\begin{aligned} \hat{H}_{\text{HC}} = & -J_x \sum_{l,m} [(e^{i2m\Phi} \hat{b}_{l+1,m}^\dagger \hat{b}_{l,m} + \lambda \hat{b}_{l,m+1}^\dagger \hat{b}_{l,m}) + \text{h.c.}] \\ & - V_x \sum_{l,m} (\hat{n}_{l,m} \hat{n}_{l+1,m} + \lambda \hat{n}_{l,m} \hat{n}_{l,m+1}) + \varepsilon_0 \sum_{l,m} \hat{n}_{l,m} \end{aligned} \quad (S.73)$$

with $V_x = \Delta_x$, $V_y = \Delta_y$ and $\varepsilon_0 = \Delta_x + \Delta_y - B_0$. Here we have removed a constant energy shift $\frac{1}{4}(2B_0 - \Delta_x - \Delta_y)L_x L_y$. Since the term $\varepsilon_0 \sum_{l,m} \hat{n}_{l,m}$ commutes with the other part of the Hamiltonian, it only causes a constant energy shift and thus can be removed from the Hamiltonian without changing the physics. Finally, our effective hard-core boson model obeys,

$$\begin{aligned} \hat{H} = & -J_x \sum_{l,m} [(e^{i2\pi\beta m} \hat{b}_{l+1,m}^\dagger \hat{b}_{l,m} + \lambda \hat{b}_{l,m+1}^\dagger \hat{b}_{l,m}) + \text{h.c.}] \\ & - V_x \sum_{l,m} (\hat{n}_{l,m} \hat{n}_{l+1,m} + \lambda \hat{n}_{l,m} \hat{n}_{l,m+1}) \end{aligned} \quad (S.74)$$

with $\beta = \Phi/\pi$ and $\lambda = J^2/K^2$.

III. COTRANSLATIONAL SYMMETRY OF TWO-MAGNON EXCITATIONS

In this section, to understand the eigenvalue problem of two-magnon excitations (two hard-core bosons), we show the cotranslational symmetry of two-magnon excitations. The two-particle Hilbert space for the model (S.74) can be spanned by the basis,

$$\mathcal{B}_{2D}^{(2)} = \{|l_1, m_1; l_2, m_2\rangle = \hat{b}_{l_1, m_1}^\dagger \hat{b}_{l_2, m_2}^\dagger |0\rangle\}, \quad (S.75)$$

with $(1 \leq l_1 < l_2 \leq L_x)$ or $(1 \leq l_1 = l_2 \leq L_x)$ and $(1 \leq m_1 < m_2 \leq L_y)$. We consider the periodic boundary conditions (PBCs) in both x - and y -directions.

Introducing $\psi_{l_1, m_1; l_2, m_2} = \langle 0 | \hat{b}_{l_2, m_2} \hat{b}_{l_1, m_1} | \Psi \rangle$, the eigenstates can be expanded as

$$|\Psi\rangle = \sum_{l_1, m_1; l_2, m_2} \psi_{l_1, m_1; l_2, m_2} |l_1, m_1; l_2, m_2\rangle. \quad (S.76)$$

The eigenequation $\hat{H} |\Psi\rangle = E |\Psi\rangle$ gives

$$\begin{aligned} E \psi_{l_1, m_1; l_2, m_2} = & -V_x (\delta_{l_2, m_2}^{l_1 \pm 1, m_1} + \lambda \delta_{l_2, m_2}^{l_1, m_1 \pm 1}) \psi_{l_1, m_1; l_2, m_2} \\ & - J_x (e^{i\Theta m_1} \psi_{l_1-1, m_1; l_2, m_2} + e^{-i\Theta m_1} \psi_{l_1+1, m_1; l_2, m_2} \\ & + e^{i\Theta m_2} \psi_{l_1, m_1; l_2-1, m_2} + e^{-i\Theta m_2} \psi_{l_1, m_1; l_2+1, m_2} \\ & + \lambda \psi_{l_1, m_1-1; l_2, m_2} + \lambda \psi_{l_1, m_1+1; l_2, m_2} \\ & + \lambda \psi_{l_1, m_1; l_2, m_2-1} + \lambda \psi_{l_1, m_1; l_2, m_2+1}) \end{aligned} \quad (S.77)$$

with $\Theta = 2\pi\beta = 2\Phi$. The PBCs require $\psi_{l_1+L_x, m_1; l_2, m_2} = \psi_{l_1, m_1; l_2+L_x, m_2} = \psi_{l_1, m_1+L_y; l_2, m_2} = \psi_{l_1, m_1; l_2, m_2+L_y} = \psi_{l_1, m_1; l_2, m_2}$, and the hard-core bosonic commutation relations require $\psi_{l_1, m_1; l_2, m_2} = \psi_{l_2, m_2; l_1, m_1}$ and $\psi_{l_1, m_1; l_1, m_1} = 0$.

To describe the cotranslational symmetry along x -direction, we introduce the two-particle cotranslational operator T_1^x along x -direction as

$$T_1^x \psi_{l_1, m_1; l_2, m_2} = \psi_{l_1+1, m_1; l_2+1, m_2}. \quad (S.78)$$

It's easy to find that $HT_1^x \psi_{l_1, m_1; l_2, m_2} = T_1^x H \psi_{l_1, m_1; l_2, m_2}$ holds for arbitrary $\psi_{l_1, m_1; l_2, m_2}$. Therefore, the Hamiltonian H commutes with T_1^x and they share a set of common eigenstates: $\psi_{l_1, m_1; l_2, m_2} = e^{\frac{i}{2}k_x(l_1+l_2)} \phi_{l_1, m_1; l_2, m_2}$, in which $\phi_{l_1+1, m_1; l_2+1, m_2} = \phi_{l_1, m_1; l_2, m_2}$ is invariant under T_1^x . The eigenequation $H_{k_x} |\Phi\rangle = E_{k_x} |\Phi\rangle$ gives

$$\begin{aligned} E_{k_x} \phi_{l_1, m_1; l_2, m_2} = & -V_x (\delta_{l_2, m_2}^{l_1 \pm 1, m_1} + \lambda \delta_{l_2, m_2}^{l_1, m_1 \pm 1}) \phi_{l_1, m_1; l_2, m_2} \\ & - J_x (e^{i\Theta m_1 - i\frac{k_x}{2}} \phi_{l_1-1, m_1; l_2, m_2} + e^{-i\Theta m_1 + i\frac{k_x}{2}} \phi_{l_1+1, m_1; l_2, m_2} \\ & + e^{i\Theta m_2 - i\frac{k_x}{2}} \phi_{l_1, m_1; l_2-1, m_2} + e^{-i\Theta m_2 + i\frac{k_x}{2}} \phi_{l_1, m_1; l_2+1, m_2} \\ & + \lambda \phi_{l_1, m_1-1; l_2, m_2} + \lambda \phi_{l_1, m_1+1; l_2, m_2} \\ & + \lambda \phi_{l_1, m_1; l_2, m_2-1} + \lambda \phi_{l_1, m_1; l_2, m_2+1}). \end{aligned} \quad (S.79)$$

Here, H_{k_x} denotes the k_x -block of the two-particle Hamiltonian and $k_x = \frac{2\pi}{L_x} \alpha_x$ is the c.o.m quasi-momentum

along x -direction (where the integer α_x satisfies $-\frac{L_x-1}{2} \leq \alpha_x \leq \frac{L_x-1}{2}$). Correspondingly, the PBCs require $\phi_{l_1+L_x, m_1; l_2, m_2} = \phi_{l_1, m_1; l_2+L_x, m_2} = (-1)^{\alpha_x} \phi_{l_1, m_1; l_2, m_2}$ and $\phi_{l_1, m_1+L_y; l_2, m_2} = \phi_{l_1, m_1; l_2, m_2+L_y} = \phi_{l_1, m_1; l_2, m_2}$, and the commutation relations require $\phi_{l_1, m_1; l_2, m_2} = \phi_{l_2, m_2; l_1, m_1}$ and $\phi_{l_1, m_1; l_1, m_1} = 0$.

Now we assume that the lattice size in the y -direction is commensurate with the rational flux caused by the synthetic gauge field. That is, $s = L_y/q$ is an integer and the rational flux is determined by $\beta = p/q$ of two coprime integer numbers p and q . To show the cotranslational symmetry along y -direction, we introduce the corresponding two-particle cotranslational operator T_q^y as

$$T_q^y \psi_{l_1, m_1; l_2, m_2} = \psi_{l_1, m_1+q; l_2, m_2+q}. \quad (\text{S.80})$$

In the k_x -subspace, it turns out to be $T_q^y \phi_{l_1, m_1; l_2, m_2} = \phi_{l_1, m_1+q; l_2, m_2+q}$. It is easy to find that $HT_q^y \psi = T_q^y H \psi$ holds for arbitrary $\psi = \psi_{l_1, m_1; l_2, m_2}$, which gives $H_{k_x} T_q^y \phi_{l_1, m_1; l_2, m_2} = T_q^y H_{k_x} \phi_{l_1, m_1; l_2, m_2}$. Therefore, H_{k_x} and T_q^y have a common set of eigenstates, which can be written as $\phi_{l_1, m_1; l_2, m_2} = e^{\frac{i}{2} k_y (m_1+m_2)} \varphi_{l_1, m_1; l_2, m_2}$, where $\varphi_{l_1, m_1+q; l_2, m_2+q} = \varphi_{l_1, m_1; l_2, m_2}$ is invariant under T_q^y . The eigenequation $H_{k_x, k_y} |\varphi\rangle = E_{k_x, k_y} |\varphi\rangle$ reads

$$\begin{aligned} E_{k_x, k_y} \varphi_{l_1, m_1; l_2, m_2} &= -V_x (\delta_{l_2, m_2}^{l_1 \pm 1, m_1} + \lambda \delta_{l_2, m_2}^{l_1, m_1 \pm 1}) \varphi_{l_1, m_1; l_2, m_2} \\ &- J_x (e^{i\Theta_{m_1} - \frac{i}{2} k_x} \varphi_{l_1-1, m_1; l_2, m_2} + e^{-i\Theta_{m_1} + \frac{i}{2} k_x} \varphi_{l_1+1, m_1; l_2, m_2} \\ &+ e^{i\Theta_{m_2} - \frac{i}{2} k_x} \varphi_{l_1, m_1; l_2-1, m_2} + e^{-i\Theta_{m_2} + \frac{i}{2} k_x} \varphi_{l_1, m_1; l_2+1, m_2} \\ &+ \lambda e^{-\frac{i}{2} k_y} \varphi_{l_1, m_1-1; l_2, m_2} + \lambda e^{\frac{i}{2} k_y} \varphi_{l_1, m_1+1; l_2, m_2} \\ &+ \lambda e^{-\frac{i}{2} k_y} \varphi_{l_1, m_1; l_2, m_2-1} + \lambda e^{\frac{i}{2} k_y} \varphi_{l_1, m_1; l_2, m_2+1}). \end{aligned} \quad (\text{S.81})$$

Here, $k_y = \frac{2\pi}{L_y} \alpha_y$ is the c.o.m quasi-momentum along y -direction (with the integer α_y satisfying $-\frac{s-1}{2} \leq \alpha_y \leq \frac{s-1}{2}$). The Eq. (S.81) gives the (k_x, k_y) -block of the Hamiltonian. By diagonalizing H_{k_x, k_y} , in addition to the continuum subband, one may find two-magnon bound-state subbands and their corresponding eigenstates. In Fig. S2(a), we show the 6 bound-state subbands for $\beta = 1/3$.

To characterize the x -type [y -type] bound-states, which are approximated by a superposition of states of the form $|lm; l+1, m\rangle$ [$|lm; l, m+1\rangle$], we introduce $P_x = \sum_{l, m} |\psi_{l, m; l+1, m}|^2$ [$P_y = \sum_{l, m} |\psi_{l, m; l, m+1}|^2$]. In Fig. S2(b)-(d), we show P_x , P_y and $P_x + P_y$ for the 6 eigen bound-states with $(k_x, k_y) = (0, 0)$ and $\lambda = 1.2 > 1$. Obviously, P_x for the three higher subbands are very close to 1, this means the eigenstates of three higher subbands are x -type bound-states, see Fig. S2(b). However, P_y for the three lowest subbands are very close to 1, this means the eigenstates of three lowest subbands are y -type bound-states, see Fig. S2(c). Moreover, all bound-states are signatured by $P = P_x + P_y \simeq 1$, see Fig. S2(d).

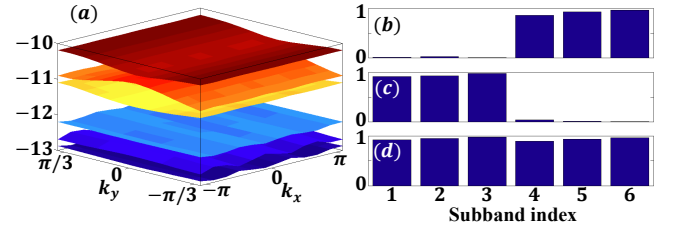


FIG. S2. (color online). (a) Two-magnon bound-state subbands for $\beta = 1/3$, $V_x/J_x = 10$, $\lambda = 1.2$, $L_y = 33$, and $L_x = 11$. Signature of bound-states for the 6 eigen bound-states with $(k_x, k_y) = (0, 0)$: (b) P_x , (c) P_y , and (d) $P = P_x + P_y$.

IV. DERIVATION OF THE EFFECTIVE SINGLE-PARTICLE MODEL FOR TWO-MAGNON BOUND-STATES

By regarding a two-magnon bound state as a quasi-particle, we analytically derive an effective single-particle model via the Schrieffer-Wolff transformation [S5]. As bound-states appear when $|V_x/J_x| \gg 1$, one may treat the hopping term

$$\hat{H}_1 = -J_x \sum_{l, m} [(e^{i2\pi\beta m} \hat{b}_{l+1, m}^\dagger \hat{b}_{l, m} + \lambda \hat{b}_{l, m+1}^\dagger \hat{b}_{l, m}) + \text{h.c.}] \quad (\text{S.82})$$

as a perturbation to the interaction term

$$\hat{H}_0 = -V_x \sum_{l, m} (\hat{n}_{l, m} \hat{n}_{l+1, m} + \lambda \hat{n}_{l, m} \hat{n}_{l, m+1}). \quad (\text{S.83})$$

Obviously, all two-magnon Fock states $|l_1, m_1; l_2, m_2\rangle = \hat{b}_{l_1, m_1}^\dagger \hat{b}_{l_2, m_2}^\dagger |0\rangle$ are eigenstates of \hat{H}_0 with eigenvalues $E_{l_1, m_1; l_2, m_2} = -V_x (\delta_{l_2, m_2}^{l_1 \pm 1, m_1} + \lambda \delta_{l_2, m_2}^{l_1, m_1 \pm 1})$. The two-magnon bound-states can be approximated by superpositions: $|G_{l, m}^x\rangle = |l, m; l+1, m\rangle$ and $|G_{l, m}^y\rangle = |l, m; l, m+1\rangle$, which are also eigenstates of \hat{H}_0 with eigenvalues $E_0^x = -V_x$ and $E_0^y = -\lambda V_x$ (where $\hat{H}_0 |G_{l, m}^x\rangle = E_0^x |G_{l, m}^x\rangle$ and $\hat{H}_0 |G_{l, m}^y\rangle = E_0^y |G_{l, m}^y\rangle$).

By implementing the SW transformation [S5], the effective single-particle Hamiltonian up to second-order reads

$$\hat{H}_{\text{eff}}^{(2)} = \hat{h}_0 + \hat{h}_2, \quad (\text{S.84})$$

$$\hat{h}_0 = -V_x (\hat{P}_1 + \lambda \hat{P}_2), \quad (\text{S.85})$$

$$\begin{aligned} \hat{h}_2 = & -\frac{1}{V_x} \left(\hat{P}_1 \hat{H}_1 \hat{H}_1 \hat{P}_1 + \frac{1}{\lambda} \hat{P}_2 \hat{H}_1 \hat{H}_1 \hat{P}_2 \right) \\ & - \frac{\lambda + 1}{2\lambda V_x} (\hat{P}_1 \hat{H}_1 \hat{H}_1 \hat{P}_2 + \hat{P}_2 \hat{H}_1 \hat{H}_1 \hat{P}_1). \end{aligned} \quad (\text{S.86})$$

Here, the two bound-state projectors are defined as

$$\begin{cases} \hat{P}_1 = \sum_{l, m} |G_{l, m}^x\rangle \langle G_{l, m}^x|, \\ \hat{P}_2 = \sum_{l, m} |G_{l, m}^y\rangle \langle G_{l, m}^y|. \end{cases} \quad (\text{S.87})$$

For convenience, we introduce the following notations

$$\begin{cases} \hat{H}_{J_x} = -J_x \sum_{l,m} (\hat{t}_{l,m}^x + \hat{t}_{l,m}^{x\dagger}), \\ \hat{H}_{J_y} = -\lambda J_x \sum_{l,m} (\hat{t}_{l,m}^y + \hat{t}_{l,m}^{y\dagger}), \end{cases} \quad (\text{S.88})$$

with

$$\begin{cases} \hat{t}_{l,m}^x = e^{i2\pi\beta m} \hat{b}_{l+1,m}^\dagger \hat{b}_{l,m}, \\ \hat{t}_{l,m}^y = \hat{b}_{l,m+1}^\dagger \hat{b}_{l,m}. \end{cases} \quad (\text{S.89})$$

It is easy to find that

$$\begin{cases} \hat{P}_1 \hat{H}_{J_x} \hat{H}_{J_y} \hat{P}_1 = \hat{P}_1 \hat{H}_{J_y} \hat{H}_{J_x} \hat{P}_1 = 0 \\ \hat{P}_2 \hat{H}_{J_x} \hat{H}_{J_y} \hat{P}_2 = \hat{P}_2 \hat{H}_{J_y} \hat{H}_{J_x} \hat{P}_2 = 0 \\ \hat{P}_1 \hat{H}_{J_x} \hat{H}_{J_x} \hat{P}_2 = \hat{P}_1 \hat{H}_{J_y} \hat{H}_{J_y} \hat{P}_2 = 0 \\ \hat{P}_1 \hat{H}_{J_x} \hat{H}_{J_y} \hat{P}_2 = \hat{P}_2 \hat{H}_{J_y} \hat{H}_{J_x} \hat{P}_1 = 0 \end{cases} \quad (\text{S.90})$$

As $\hat{H}_1 = \hat{H}_{J_x} + \hat{H}_{J_y}$, we have

$$\begin{cases} \hat{P}_1 \hat{H}_1 \hat{H}_1 \hat{P}_1 = \hat{P}_1 \hat{H}_{J_x} \hat{H}_{J_x} \hat{P}_1 + \hat{P}_1 \hat{H}_{J_y} \hat{H}_{J_y} \hat{P}_1 \\ \hat{P}_2 \hat{H}_1 \hat{H}_1 \hat{P}_2 = \hat{P}_2 \hat{H}_{J_x} \hat{H}_{J_x} \hat{P}_2 + \hat{P}_2 \hat{H}_{J_y} \hat{H}_{J_y} \hat{P}_2 \\ \hat{P}_1 \hat{H}_1 \hat{H}_1 \hat{P}_2 = \hat{P}_1 \hat{H}_{J_y} \hat{H}_{J_x} \hat{P}_2 \\ \hat{P}_2 \hat{H}_1 \hat{H}_1 \hat{P}_1 = \hat{P}_2 \hat{H}_{J_x} \hat{H}_{J_y} \hat{P}_1. \end{cases} \quad (\text{S.91})$$

By using the hard-core bosonic commutation relations, one can obtain

$$\begin{cases} \hat{H}_{J_x} |G_{l,m}^x\rangle = -J_x \left(e^{i2\pi\beta m} \hat{b}_{l,m}^\dagger \hat{b}_{l+2,m}^\dagger + e^{-i2\pi\beta m} \hat{b}_{l-1,m}^\dagger \hat{b}_{l+1,m}^\dagger \right) |0\rangle \\ \hat{H}_{J_x} |G_{l,m}^y\rangle = -J_x \left(e^{i2\pi\beta m} \hat{b}_{l,m+1}^\dagger \hat{b}_{l+1,m}^\dagger + e^{i2\pi\beta(m+1)} \hat{b}_{l,m}^\dagger \hat{b}_{l+1,m+1}^\dagger + e^{-i2\pi\beta m} \hat{b}_{l-1,m}^\dagger \hat{b}_{l,m+1}^\dagger + e^{-i2\pi\beta(m+1)} \hat{b}_{l-1,m+1}^\dagger \hat{b}_{l,m}^\dagger \right) |0\rangle \\ \hat{H}_{J_y} |G_{l,m}^x\rangle = -\lambda J_x \left(\hat{b}_{l,m+1}^\dagger \hat{b}_{l+1,m}^\dagger + \hat{b}_{l,m}^\dagger \hat{b}_{l+1,m+1}^\dagger + \hat{b}_{l,m-1}^\dagger \hat{b}_{l+1,m}^\dagger + \hat{b}_{l,m}^\dagger \hat{b}_{l+1,m-1}^\dagger \right) |0\rangle \\ \hat{H}_{J_y} |G_{l,m}^y\rangle = -\lambda J_x \left(\hat{b}_{l,m}^\dagger \hat{b}_{l,m+2}^\dagger + \hat{b}_{l,m-1}^\dagger \hat{b}_{l,m+1}^\dagger \right) |0\rangle. \end{cases} \quad (\text{S.92})$$

Therefore, we get

$$\begin{cases} \hat{P}_1 \hat{H}_{J_x} \hat{H}_{J_x} \hat{P}_1 = J_x^2 \sum_{l,m} \left(e^{i4\pi\beta m} |G_{l+1,m}^x\rangle \langle G_{l,m}^x| + \text{h.c.} + 2 |G_{l,m}^x\rangle \langle G_{l,m}^x| \right) \\ \hat{P}_1 \hat{H}_{J_y} \hat{H}_{J_y} \hat{P}_1 = 2\lambda^2 J_x^2 \sum_{l,m} \left(|G_{l,m+1}^x\rangle \langle G_{l,m}^x| + \text{h.c.} + 2 |G_{l,m}^x\rangle \langle G_{l,m}^x| \right) \\ \hat{P}_2 \hat{H}_{J_x} \hat{H}_{J_x} \hat{P}_2 = 2J_x^2 \sum_{l,m} \left(e^{i2\pi\beta(2m+1)} |G_{l+1,m}^y\rangle \langle G_{l,m}^y| + \text{h.c.} + 2 |G_{l,m}^y\rangle \langle G_{l,m}^y| \right) \\ \hat{P}_2 \hat{H}_{J_y} \hat{H}_{J_y} \hat{P}_2 = \lambda^2 J_x^2 \sum_{l,m} \left(|G_{l,m+1}^y\rangle \langle G_{l,m}^y| + \text{h.c.} + 2 |G_{l,m}^y\rangle \langle G_{l,m}^y| \right) \end{cases} \quad (\text{S.93})$$

and

$$\begin{aligned} & \hat{P}_1 \hat{H}_{J_y} \hat{H}_{J_x} \hat{P}_2 + \hat{P}_2 \hat{H}_{J_x} \hat{H}_{J_y} \hat{P}_1 \\ &= 2\lambda J_x^2 \cos(\pi\beta) \sum_{l,m} \left[e^{i2\pi\beta m} e^{i\pi\beta} \left(|G_{l,m}^x\rangle \langle G_{l,m}^y| + |G_{l+1,m}^y\rangle \langle G_{l,m}^x| + |G_{l,m+1}^x\rangle \langle G_{l,m}^y| + |G_{l+1,m}^y\rangle \langle G_{l,m+1}^x| \right) + \text{h.c.} \right]. \end{aligned} \quad (\text{S.94})$$

Insert Eqs. (S.91), (S.93), (S.94) into Eq. (S.86), we obtain

$$\begin{aligned} \hat{H}_{\text{eff}}^{(2)} = & -J_{\text{eff}} \sum_{l,m} \left\{ \left[e^{i4\pi\beta m} |G_{l+1,m}^x\rangle \langle G_{l,m}^x| + 2\lambda^2 |G_{l,m+1}^x\rangle \langle G_{l,m}^x| + \frac{2}{\lambda} e^{i4\pi\beta m} e^{i2\pi\beta} |G_{l+1,m}^y\rangle \langle G_{l,m}^y| + \lambda |G_{l,m+1}^y\rangle \langle G_{l,m}^y| + J_{xy} e^{i2\pi\beta m} e^{i\pi\beta} \left(|G_{l,m}^x\rangle \langle G_{l,m}^y| + |G_{l+1,m}^y\rangle \langle G_{l,m}^x| + |G_{l,m+1}^x\rangle \langle G_{l,m}^y| + |G_{l+1,m}^y\rangle \langle G_{l,m+1}^x| \right) \right] + \text{h.c.} \right. \\ & \left. + \epsilon_x |G_{l,m}^x\rangle \langle G_{l,m}^x| + \epsilon_y |G_{l,m}^y\rangle \langle G_{l,m}^y| \right\}. \end{aligned} \quad (\text{S.95})$$

Here, $J_{\text{eff}} = J_x^2/V_x$, $J_{xy} = (\lambda+1) \cos(\pi\beta)$, $\epsilon_x = V_x^2/J_x^2 + 2 + 4\lambda^2$, and $\epsilon_y = \lambda V_x^2/J_x^2 + 2\lambda + 4/\lambda$.

In order to capture the single-particle nature of the bound-states, we introduce the creation operators $\hat{A}_{l,m}^\dagger$ and $\hat{B}_{l,m}^\dagger$ as follows: $\hat{A}_{l,m}^\dagger$ creates a quasi-particle in the x -type bound-state $|G_{l,m}^x\rangle$, while $\hat{B}_{l,m}^\dagger$ creates a quasi-particle in the y -type bound-state $|G_{l,m}^y\rangle$. That is, we define a mapping between two-magnon bound-states and single-particle states: $|G_{l,m}^x\rangle \Leftrightarrow \hat{A}_{l,m}^\dagger |0\rangle$ and $|G_{l,m}^y\rangle \Leftrightarrow \hat{B}_{l,m}^\dagger |0\rangle$. Thus the effective single-particle Hamiltonian (S.95) becomes

$$\begin{aligned} \hat{H}_{\text{eff}} = & -J_{\text{eff}} \sum_{lm} \left\{ \left[e^{i4\pi\beta m} \hat{A}_{l+1,m}^\dagger \hat{A}_{l,m} + 2\lambda^2 \hat{A}_{l,m+1}^\dagger \hat{A}_{l,m} + \frac{2}{\lambda} e^{i4\pi\beta m} e^{i2\pi\beta} \hat{B}_{l+1,m}^\dagger \hat{B}_{l,m} + \lambda \hat{B}_{l,m+1}^\dagger \hat{B}_{l,m} + J_{xy} e^{i2\pi\beta m} e^{i\pi\beta} \left(\hat{A}_{l,m}^\dagger \hat{B}_{l,m} + \hat{B}_{l+1,m}^\dagger \hat{A}_{l,m} + \hat{A}_{l,m+1}^\dagger \hat{B}_{l,m} + \hat{B}_{l+1,m}^\dagger \hat{A}_{l,m+1} \right) \right] + \text{h.c.} \right. \\ & \left. + \epsilon_x \hat{A}_{l,m}^\dagger \hat{A}_{l,m} + \epsilon_y \hat{B}_{l,m}^\dagger \hat{B}_{l,m} \right\}, \end{aligned} \quad (\text{S.96})$$

which describes a Hofstadter superlattice with two coupled standard Hofstadter lattices A and B .

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